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Letter

Development and Application of a Polydimethylsiloxane-Based Passive Air Sampler to Assess Personal Exposure to SARS-CoV-2

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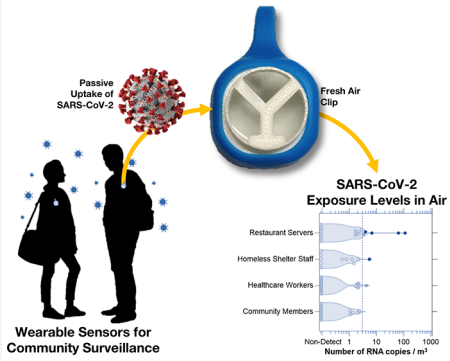
ABSTRACT: Exhaled respiratory droplets and aerosols can carry infectious viruses and are an important mode of transmission for COVID-19. Recent studies have been successful in detecting airborne SARS-CoV-2 RNA in indoor settings using active sampling methods. The cost, size, and maintenance of these samplers, however, limit their long-term monitoring ability in high-risk transmission areas. As an alternative, passive samplers can be small, lightweight, and inexpensive and do not require electrical power or maintenance for continual operation. Integration of passive samplers into wearable designs can be used to better understand personal exposure to the respiratory virus. This study evaluated the use of a polydimethylsiloxane (PDMS)-based passive sampler to assess personal exposure to aerosol and droplet SARS-CoV-2. The rate of uptake of virus-laden aerosol on PDMS was determined in lab-based rotating drum experiments to estimate time-weighted averaged airborne viral concentrations from passive sampler viral loading. The passive sampler was then embedded in a wearable clip design and distributed to community members across Connecticut to surveil personal SARS-CoV-2 exposure. The virus was detected on clips worn by five of the 62 participants (8%) with personal exposure ranging from 4 to 112 copies of SARS-CoV-2 RNA/m^3, predominantly in indoor restaurant settings. Our findings demonstrate that PDMS-based passive samplers may serve as a useful exposure assessment tool for airborne viral exposure in real-world high-risk settings and provide avenues for early detection of potential cases and guidance on site-specific infection control protocols that preempt community transmission.



INTRODUCTION

COVID-19, caused by severe acute respiratory syndrome coronavirus 2 (SARS-CoV-2),[1] was declared a global pandemic by the World Health Organization in March 2020,[2] with more than 263.5 million confirmed cases and 5.2 million deaths worldwide to date.[3,4] Inhalation of virus-laden aerosols and contact with respiratory droplets[5−7] that are expelled from infected individuals (asymptomatic, presymptomatic, and symptomatic)[8] during coughing, sneezing, speaking, or breathing are central routes of transmission for SARS-CoV-2.[2,9−12] Mitigating the spread of SARS-CoV-2 and other airborne respiratory viruses requires layered infection prevention and control strategies,[5,9] including the availability of clinical testing, use of masks, distancing, hand hygiene, environmental cleaning, and enhanced ventilation.[2,11]

The effectiveness of many of the infection prevention and control measures mentioned above can be evaluated using monitors that measure airborne levels of virus. Recent studies have been successful in detecting airborne SARS-CoV-2 RNA in indoor settings using active sampling methods.[13−16] However, the cost, size, and maintenance of these samplers limit their long-term monitoring ability in high-risk transmission areas, including hospital wards, nursing homes, schools, and restaurants.[17,18] Moreover, the nonportable nature of many active samplers limits their feasibility as a wearable device for evaluating personal exposures.[13−15,17,18] As an alternative, passive samplers can be small, lightweight, and inexpensive and do not require electrical power or maintenance for continual operation.[17,18] The broad integration of passive samplers in wearable designs can be used to better understand personal exposure to respiratory virus. While passive sampling is promising from a deployment perspective, it does provide additional challenges, including uncertainties with regard to aerosol uptake conditions[18] and higher detection limits[17] compared to active sampling.

This study uses a wearable passive air sampler, known as the Fresh Air Clip, to monitor personal exposure to airborne

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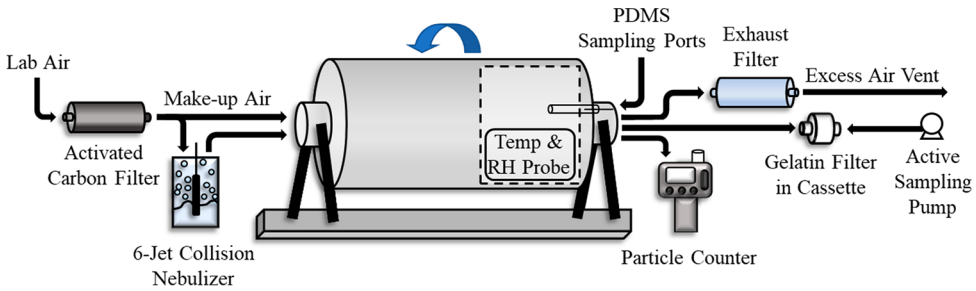

Figure 1. Experimental setup for testing the uptake of viral aerosols by a PDMS air sampler in a rotating drum. A 44.5 L aluminum drum was rotated at a constant speed. Filtered air was directly routed into the drum to supply active sampler makeup air or through a pressure gauge into a 6-Jet Collison Nebulizer to generate aerosols. Aerosol was sampled on the opposite side of the drum. Sampling ports included four retractable lines fit with 2.5 cm long PDMS sorbent tubes for passive sampling. Additional ports were used for active air sampling and real-time particle monitoring.

SARS-CoV-2. The Fresh Air Clip is a low-cost and lightweight device composed of polydimethylsiloxane (PDMS), which has previously been used to evaluate individual exposure to hydrophobic chemical contaminants that are absorbed by the polymeric surface.[19−21] Additional studies have also used PDMS as a model surface for salivary protein adsorption[22] and demonstrated its ability to efficiently capture nonpolar compounds,[19,23] such as lipid-enveloped viruses. In this study, we employed the use of a rotating drum[24,25] to investigate the rate of uptake of virus-laden aerosol on PDMS to estimate time-weighted average airborne viral concentrations from passive sampler viral loading results. Fresh Air Clips then were distributed to community members across Connecticut to surveil personal SARS-CoV-2 exposure. Monitoring airborne SARS-CoV-2 with wearable sampling devices could facilitate risk assessments for virus transmission, providing avenues for early detection of potential cases and guidance on site-specific infection control protocols that preempt community transmission.

■ MATERIALS AND METHODS

Experimental Uptake Rate Determination. *Viral Surrogate Φ6.* The bacteriophage Phi6 (Φ6) was used as a BSL-1 surrogate organism to estimate the rate of uptake of virus-laden aerosols by PDMS.[10,26,27] Φ6 has previously been explored as a surrogate for various enveloped viruses in environmental exposure and persistence studies[10,28−30] and was recently utilized as a surrogate for SARS-CoV-2 because its physiological characteristics are similar to those of the virus, including a diameter ranging from 75 to 100 nm (vs a diameter of 90−110 nm for SARS-CoV-2),[31] a spherical shape with protruding spike proteins, a lipid envelope, and an RNA genome.[10,26,32,33]

Rotating Drum Configuration, Aerosol Generation, and Aerosol Sampling. A custom aluminum drum was constructed to determine the rate of uptake of virus-laden aerosols by PDMS (Figure 1). The drum rotated at a constant speed of 2.9 rpm to minimize aerosol loss.[34] Laboratory air was routed through an activated carbon filter to provide purified air to a nebulizer (BGI Inc. 6-Jet Collison Nebulizer) and to supply makeup due to losses from active samplers. The drum was maintained at 20 °C throughout the tests. The phage lysate in artificial saliva [∼1.9×10^5 gene copies of Φ6/μL (Table S1)] was nebulized at 20 psi for 10 s to generate polydisperse Φ6-containing aerosols. To better simulate the size of virus-laden aerosols released by infected people through various respiratory activities,[35] $CaCl_2$ (0.25 M) was added to the nebulizing solution as a coagulant to promote aerosol agglomeration and increased the rate of generation of larger aerosols (1.0−5.0 μm). Typical size distributions of the generated aerosols are shown in Figure S1.

Passive sampling, active sampling, and real-time size-resolved aerosol measurements were performed 3 min after aerosolization to ensure a homogeneous distribution of aerosol in the drum. For passive sampling, 2.5 cm long PDMS sorbent tubes (effective sampling area of 0.09 cm^2; SILASTIC Laboratory tubing) were inserted into the drum following aerosolization and were removed after various sampling periods (ranging from 30 min to 2 h). Triplicate PDMS sorbent tube samples were collected for each exposure duration. For active sampling, air was sampled from the drum through a gelatin filter (Sartorius Stedim Biotech) mounted in a filter cassette (SKC Ltd.). Active air samples were collected every 30 min at a rate of 1.5 L/min for 2 min over the 2 h test periods. The aerosol number concentration was monitored before and after active sampling using a size-resolved (<0.3, 0.3−0.5, 0.5−1.0, 1.0−2.5, 2.5−5.0, and 5.0−10 μm) optical particle counter (MET ONE HHPC-6 Airborne Particle Counter), with an average of two measurements used as the aerosol concentration at that time point. Passive and active air samples collected from uptake experiments were stored in microcentrifuge tubes at −80 °C prior to extraction and quantification of viral RNA. Replicate uptake rate experiments were conducted.

Calculating the Rate of Uptake of Viral Aerosols on PDMS. To estimate airborne viral time-averaged concentrations using PDMS passive air samplers, the rate of uptake of virus-laden aerosols by this sorbent was determined on the basis of active measurements. This uptake rate (R), expressed as cubic meters of air sampled per hour per square centimeter of PDMS, was derived as follows:

$$R = \frac{m_{\mathrm{RNA}}}{\overline{C}_{\mathrm{RNA}} t} = \frac{m_{\mathrm{RNA}}}{C_{\mathrm{RNA\_PM}} \overline{C}_{\mathrm{PM}} t} \tag{1}$$

where m_{RNA} (RNA copies/cm^2 of PDMS) denotes the viral RNA loading on a unit area of PDMS, which was back-calculated on the basis of the recovered RNA quantities and the recovery of the virus from PDMS [131 ± 19% and 45 ± 8% for Φ6 and SARS-CoV-2, respectively (Figure S2)], and $\overline{C}_{\mathrm{RNA}}$ (RNA copies/m^3) is the time-weighted average virus concentration in the drum air over the sampling duration t (hours). The denominator $\overline{C}_{\mathrm{RNA}} t$, as a whole, is a measure of the cumulative exposure to virus during the sampling period,[36] and it was calculated by multiplying the Φ6 concentration contained in the aerosols ($C_{\mathrm{RNA\_PM}}$, copies per microgram of

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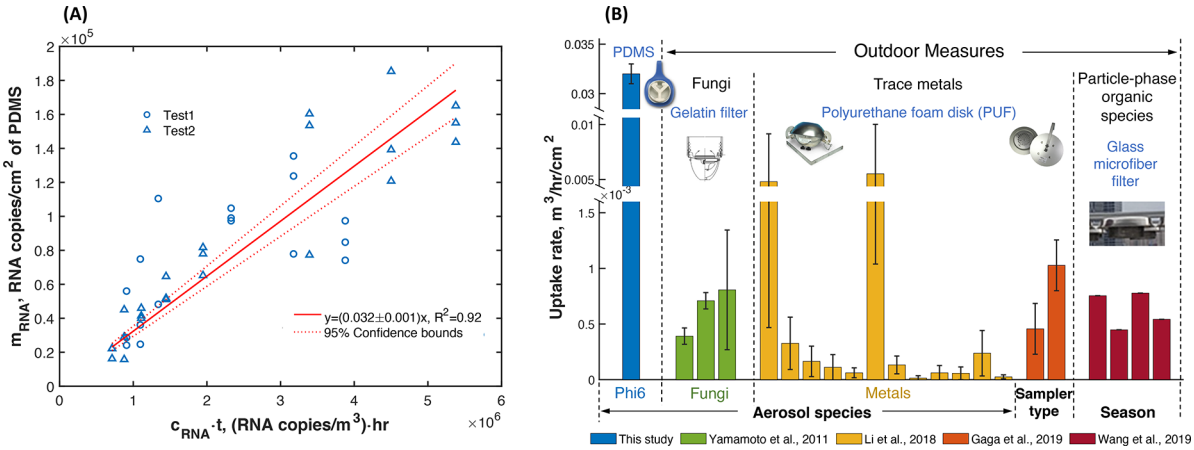

Figure 2. (A) PDMS uptake of Φ6 as a function of cumulative virus exposure. The triplicate measurements from the two experimental tests are displayed. The linear regression was fitted using all individual replicates (N = 41), and the dashed lines indicate the 95% confidence intervals. (B) Uptake rates of passive samplers for airborne aerosol species measured by a rotating drum in this work (bacteriophage) and reported in the field studies for various fungal aerosols,[40] trace metals,[41,42] and particle-phase persistent organic species.[43] The uptake rates were plotted against a variety of affecting factors (aerosol species, sampler type or configuration, season, etc.) after being normalized on the basis of the effective collection area of passive samplers. Error bars represent measurement uncertainties.

aerosol) by the cumulative PDMS exposure to viral aerosols ($\overline{C}_{PM}t$, μg m^{-3} h).

C_{RNA_PM} was determined from active samples:

$$C_{RNA\_PM} = \frac{m_{RNA\_filter}}{C_{PM\_sampling} V_{air}} \tag{2}$$

where m_{RNA_filter} (copies) is the RNA amount measured from the gelatin filter, $C_{PM\_sampling}$ (μg m^{-3} of air) is the average mass concentration of suspended aerosol in the drum air during active sampling, and V_{air} (m^3) is the air volume passed through the gelatin filter. Assuming first-order aerosol decay in the drum, the cumulative aerosol exposure by PDMS was calculated as the sum of time-integrated mass concentrations of aerosols with different sizes:

$$\overline{C}_{PM}t = \sum_{i=1}^{5} \int_{t_1}^{t_2} C_{PM0\_i} e^{-k_i t}\, dt \tag{3}$$

where $\overline{C}_{PM}$ (μg m^{-3}) is the time-weighted average mass concentration of aerosols suspended in the drum air during $t = t_1 \sim t_2$, C_{PM0_i} (μg m^{-3}) is the initial concentration of aerosol in the ith size bin, and k_i is its corresponding decay constant. The decay constants for aerosols with different sizes were calculated on the basis of particle counter measurements (Figure S3). The mass concentrations of aerosol were converted from number concentrations assuming spherical aerosols and unity aerosol density (i.e., ρ = 1 g/cm^3). Detailed calculations are described in the Supporting Information.

The calculated uptake rate (R), the number of viral copies per square centimeter of PDMS (m_{RNA}), and the Fresh Air Clip sampling duration (t) were then used to estimate the time-weighted average viral aerosol concentration ($\overline{C}_{RNA}$) or the personal level of exposure to airborne virus over the assessment period ($\overline{C}_{RNA}t$) in accordance with eq 1.

Assessment of Personal Exposure to Airborne SARS-CoV-2 Using the Fresh Air Clip. A PDMS pad (4.10 cm^2 effective sampling area) was fabricated (Dow Sylgar 184 Silicone Encapsulant Clear Kit) and embedded in a three-dimensionally printed acrylonitrile butadiene styrene (ABS) chamber. A perforated cover was also three-dimensionally printed from ABS, placed over the PDMS-containing chamber, and mounted in a magnetic clip. This wearable passive air sampler design was termed the Fresh Air Clip. Additional details are provided in the Supporting Information.

Fresh Air Clips were deployed to individuals across Connecticut between January and May 2021 with participants residing in communities with high COVID-19 transmission rates or working in high-risk indoor occupational environments, such as restaurants offering indoor dining, a homeless shelter, and healthcare facilities. To capture exposure of the breathing zone (i.e., the area near the mouth and nose)[37] and allow for sufficient sampling of exposure event opportunities for airborne virus detection, the study participants wore Fresh Air Clips on their shirt collars for 5 days. Community members living in regions with high COVID-19 transmission wore the Fresh Air Clip during their daily activities (i.e., work from home, exercise, shopping, etc.). Occupational sampling was performed only while study participants were at work. Study participants placed the passive samplers in sealed plastic bags while asleep (community members) or while not at work (occupational). Participants were instructed to wear the Fresh Air Clips during their normal workday or daily activities for 5 days and completed a Qualtrics survey detailing the dates, duration, and location the Fresh Air Clip were worn as well as their activities during the sampling period. A total of 62 Fresh Air Clips were collected from study participants. The PDMS passive samplers were stored individually at −80 °C after collection. Approval for this study was obtained by the Institutional Review Board at Yale University (HIC 2000026109).

Virus Quantification. Φ6 RNA was quantified for samples (PDMS sorbent tubes and gelatin filters) collected from rotating drum experiments. SARS-CoV-2 RNA concentrations were determined for the Fresh Air Clips collected from study participants. Viral RNA was extracted from each sample type (Quick-RNA Viral Kit, Zymo Research) and quantified by droplet digital polymerase chain reaction (ddPCR), with corresponding primers and probes,[30,38,39] using the One-Step RT-ddPCR Advanced Kit for Probes (Bio-Rad). Thermocycling was performed according to the manufacturer's

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Environ. Sci. Technol. Lett. XXXX, XXX, XXX−XXX

Table 1. Location-Based Sampling of Distributed PDMS Fresh Air Clip Passive Samplers, Including Relevant SARS-CoV-2 Infection Rates[48,49]

Exposure assessment location	Number of samplers distributed	Number of samplers returned	Assessment period	Mask mandate	Averaged estimated SARS-CoV-2 daily case rate (cases per 100,000 people)
Restaurants	47	19	March to May	No (while patrons were eating)	27.2
Healthcare facilities	46	17	January to April	Yes	39.5
community	24	15	March to May	Varied	26.2
homeless shelter	26	11	March to May	Yes	19.0

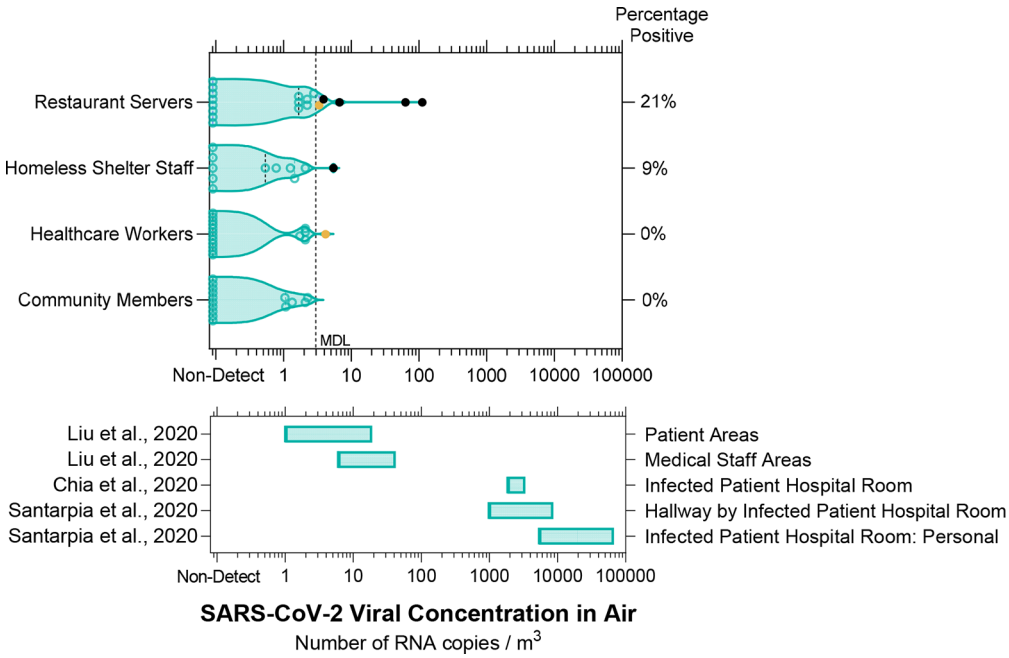

Figure 3. Distribution of SARS-CoV-2 RNA concentrations in indoor air based on PDMS Fresh Air Clip passive sampling by sampling location compared to previously reported SARS-CoV-2 RNA concentrations in indoor air using active sampling methods.[14,53,54] Black circles indicate samples that were deemed positive for SARS-CoV-2 (above the MDL with both replicates positive). Yellow circles depict samples that were above the MDL but only one replicate was positive. Thus, the sample was not counted as positive for SARS-CoV-2. The hollow blue samples report levels of SARS-CoV-2 that fell below the MDL. The percentages specify the percentage of samples per sampling location that were positive for SARS-CoV-2.

recommended protocol with annealing/extension temperatures of 60 °C for Φ6 and 55 °C for SARS-CoV-2 samples. Details can be found in the Supporting Information.

RESULTS AND DISCUSSION

Uptake of Virus-Laden Aerosols by PDMS. The kinetic uptake of virus-laden aerosols by PDMS was evaluated for cumulative exposure to bulk aerosol ranging from 80 to 600 μg m^{-3} h (Figure S4A) with a viral load of 6.3−9.0 × 10^3 RNA copies/μg of aerosol (Figure S5). A positive linear relationship was found between the RNA copies sampled by the PDMS and the cumulative Φ6 exposure (Figure S4B), suggesting the PDMS had not approached its equilibrium uptake capacity during the exposure period. The high aerosol exposure of 600 μg m^{-3} h (Figure S4A), therefore, can be used to estimate the time over which linear uptake was expected on the basis of the ambient aerosol levels in different environments. Within the linear uptake regime, the average viral concentration in ambient air over the sampling period can be quantified given the passive sampler's uptake rate.

The uptake rates determined from the two experimental tests were found to be similar (p = 0.54 for a regression comparison t test). Upon combination of all of the individual observations, the average rate of uptake of Φ6 by unit area of PDMS was determined to be 0.032 ± 0.001 m^3 h^{-1} cm^{-2} [R^2 = 0.92, and N = 41 (Figure 2A)]. This experimentally derived uptake rate is higher than the rates reported in previous studies (0.0007−0.004 m^3 h^{-1} cm^{-2}) that were derived on the basis of outdoor measurements of airborne fungi,[40] trace metals,[41,42] and persistent organic species in aerosols[43] utilizing different passive sampler configurations and collection media (Figure 2B). The variability in the uptake rates may be due to changes in aerosol composition, aerosol size distribution, sampler types, and environmental conditions (e.g., wind speed).[44−46] The sheltered design of passive air samplers used in previously published studies served to minimize variable air flow over the sorbent material by controlling the boundary layer of air above the sampling surface.[47] While this design limited variability in the rate of uptake of airborne contaminants, the rate of uptake was also reduced. To enhance aerosol deposition for our wearable passive air sampler, we used an open-face design that

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Environ. Sci. Technol. Lett. XXXX, XXX, XXX−XXX

allowed for increased air flow over the PDMS pad. The hydrophobic and porous properties of PDMS likely also enhanced uptake of virus-laden aerosol.

Assessment of Personal Exposure to Airborne SARS-CoV-2 Using the Fresh Air Clip. Sixty-two Fresh Air Clips were returned and assessed: 47 from occupational environments and 15 from community members (Table 1). The uneven distribution of total samplers analyzed per category was due to difficulties of public reliance, particularly in wearing the Fresh Air Clip for 5 days, completing the associated survey, and returning the used clip. While the Fresh Air Clip itself is easily deployable, the sampling duration of 5 days in addition to the necessity of an extended time commitment filling out the survey made it challenging for participants, particularly essential workers in high-stress environments, to reliably complete the sampling process.

We were able to reliably detect samples positive for SARS-CoV-2 with ≥6 copies of viral RNA per sampler extraction, based on analysis of method blanks (detailed in the Supporting Information). Eight percent of Fresh Air Clips were positive for SARS-CoV-2 viral RNA, with values ranging from 7 to 200 copies per clip (Figure S7). This represents the total SARS-COV-2 viral RNA detected on the passive sampler using ddPCR methods; infectious viral concentrations were not assessed. The SARS-CoV-2 occurrence observed with passive sampling is consistent with prior studies utilizing active sampling methods that reported no detection of SARS-CoV-2 among many air samples, specifically in hospital settings.[13,50] Of the positive Fresh Air Clips, four were worn by restaurant servers and one was worn by a homeless shelter staff person. Notably, two positive samples collected in restaurants with indoor dining were found to have high viral load when compared to the other samples (>100 copies per clip), suggestive of close contact with one or more infected individuals. Sampling was conducted when case rates in the communities studied ranged from ∼4 to 102 estimated daily COVID-19 cases per 100,000 people.[48] All locations were under mask mandates during the sampling; however, restaurant patrons are not required to wear masks while seated, potentially accounting for the more frequent and higher SARS-CoV-2 values observed in restaurants. Similarly, the lack of SARS-CoV-2 detection in healthcare facilities is fairly expected, as hospitals have strict personal protective equipment (PPE) requirements, cleaning protocols, and high ventilation rates that have previously been associated with decreased transmission.[2,7,51,52]

Viral load measurements on positive samplers were converted to the cumulative exposure (copies m^{-3} h) of corresponding participants to SARS-CoV-2 during deployment (eq 1), based on the viral uptake rate and the sampler collection area (the results are listed in Table S2). Ambient SARS-CoV-2 concentrations in indoor settings were further determined on the basis of the passive sampling duration of each participant (Figure 3). Positive samples estimate a range of 4−112 copies of SARS-CoV-2 RNA/m^3 (Table S2). The ambient viral levels in this study are comparable to those determined with active gelatin filter sampling in a medical staff area but lower than the levels observed in hospital rooms of infected patients.[14,15,53]

Detection of SARS-CoV-2 using Fresh Air Clips demonstrates that exposure to airborne or droplet virus can be detected using passive sampling methods. The collection of 14 copies of SARS-CoV-2 viral RNA was necessary on a Fresh Air Clip to identify a positive sampler. This is ∼21 times lower than the estimated inhalation dose for SARS-CoV-2;[55] thus, the Fresh Air Clip can detect exposure events at subinfectious doses. While the size of the study population limited comparison between microenvironments, one can conclude that PDMS passive samplers can serve as a semiquantitative screening tool for assessing personal exposure to viral aerosols. Scaling the deployment of Fresh Air Clips could facilitate the identification of high-risk areas for indoor SARS-CoV-2 exposure. More broadly, future research could apply use of this PDMS passive air sampling tool for public health situational awareness for the presence of other biological threats to the health of the public.[56]

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.1c00877.

Method details of the rotating drum experiments used to evaluate uptake of virus by the passive air sampler (Figures S1−S6 and Table S1), a comprehensive description of viral quantification and recovery methods, a visual representation of the Fresh Air Clip sampler (Figures S6 and S7 and Table S2), and additional results detailing viral loading and exposure levels for the study population (PDF)

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Notes

The authors declare no competing financial interest.

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