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Increased Indoor Exposure to Commonly Used Disinfectants during the COVID-19 Pandemic

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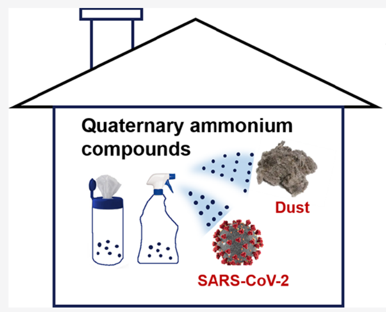
ABSTRACT: Quaternary ammonium compounds (QACs or "quats") make up a class of chemicals used as disinfectants in cleaning and other consumer products. While disinfection is recommended for maintaining a safe environment during the COVID-19 pandemic, the increased use of QACs is concerning as exposure to these compounds has been associated with adverse effects on reproductive and respiratory systems. We have determined the occurrence of 19 QACs in residential dust collected before and during the COVID-19 pandemic. QACs were detected in >90% of the samples collected during the pandemic at concentrations ranging from 1.95 to 531 μg/g (n = 40; median of 58.9 μg/g). The total QAC concentrations in these samples were significantly higher than in samples collected before the COVID-19 pandemic (p < 0.05; n = 21; median of 36.3 μg/g). Higher QAC concentrations were found in households that generally disinfected more frequently (p < 0.05). Disinfecting products commonly used in these homes were analyzed, and the QAC profiles in dust and in products were similar, suggesting that these products can be a significant source of QACs. Our findings indicate that indoor exposure to QACs is widespread and has increased during the pandemic.



■ INTRODUCTION

The spread of SARS coronavirus 2 (SARS-CoV-2), which causes the disease COVID-19, has resulted in a surge in disinfectant use to keep the indoor environment safe.[1,2] Intensified cleaning protocols during the COVID-19 pandemic specifically call for the increased use of disinfectants in homes and high-risk public spaces, such as schools, health and other care facilities, and food service and work spaces.

Disinfecting products containing quaternary ammonium compounds (QACs), also termed "quats", are recommended by the United States Centers for Disease Control and Prevention (CDC) and Environmental Protection Agency (EPA) for disinfecting procedures specifically targeting SARS-CoV-2.[3] QACs make up the major class of disinfectants and antimicrobials used in cleaning products, biocides, personal care products, and biomedical materials.[4,5] QACs are salts of quaternary ammonium cations with at least one long hydrophobic hydrocarbon-chain substituent and other short-chain substituents, such as methyl or benzyl groups. The three most widely used QAC groups include benzylalkyldimethylammonium compounds (BACs, with C6−C18 alkylated chains), dialkyldimethylammonium compounds (DDACs, with C8−C18 alkylated chains), and alkyltrimethylammonium compounds (ATMACs, with C8−C18 alkylated chains) (Figure S1). Some QACs, including C12-BAC and C14-BAC, are classified by the EPA as high-production volume chemicals based on the manufactured or imported amount exceeding 1 million pounds per year.[2] These compounds can disrupt the adipose cell membranes of living organisms and thus the viral envelopes and remove organic material. It is this property in particular that enables QACs to act as disinfectants and antimicrobials.[6]

Exposure to QACs has been associated with several adverse health effects. QACs are recognized as asthmagens, as previous animal and occupational studies have demonstrated that exposure to QACs may lead to a significant increase in asthma triggers and other breathing problems, such as pulmonary cell damage and inflammation.[7,8] Skin irritation and decreased fertility were observed in rodents and guinea pigs exposed to some QACs through inhalation and diet.[9−13] In addition, QACs increase the permeability of outer membranes of living organisms and their long-term use may disrupt the protective lipid membranes of the skin and potentially increase the absorption of toxic substances.[14] Hence, the increased use of household disinfectants and other cleaning agents containing QACs during the COVID-19 pandemic is of significant concern.[2]

QACs have been detected in wastewater sludge, surface waters, sediments, and soil.[5,15−20] Moreover, high levels of QACs have been reported in fruits, food additives, milk, and other dairy products.[21−25] However, comprehensive studies of their occurrence in the indoor environment are lacking. Indoor

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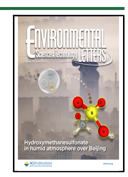

Table 1. Detection Frequencies (DF, percent), Minimum (min), Maximum (max), and Median Concentrations (micrograms per gram) of QACs in Residential Dust Collected before (n = 21) and during (n = 40) the COVID-19 Pandemic, Contributions (contr, percent) of Each QAC to the ∑QAC Concentrations, and Percent Changes in Concentrations Measured in These Two Sample Groups (based on median concentrations)[a]

	before COVID-19					during COVID-19					
QACs	DF	min	max	median	contr	DF	min	max	median	contr	change (%)
BACs											
C6-BAC	95	<MDL	0.015	0.00171	0.01	98	<MDL	0.084	0.004	0.01	134*
C8-BAC	100	0.0022	12.2	0.0496	0.2	100	0.0022	7.58	0.058	0.1	17
C10-BAC	100	0.0040	0.329	0.0213	0.1	100	0.0005	0.787	0.054	0.1	154*
C12-BAC	100	1.40	32.5	5.89	25	100	0.244	181	12.6	29	114*
C14-BAC	100	0.863	30.9	3.88	16	100	0.760	154	9.55	22	146*
C16-BAC	100	0.181	9.73	1.03	4.3	100	0.203	75.6	3.17	7.2	208*
C18-BAC	100	0.0393	6.07	0.431	1.8	100	0.061	34.8	1.16	2.6	169*
∑BAC	100	3.19	74.2	14.2	48	100	1.66	421	27.1	56	91*
DDACs											
C8-DDAC	100	0.056	7.33	1.10	4.6	100	0.0148	20.2	1.63	3.7	48
C10-DDAC	100	1.09	24.1	5.53	23	100	0.0219	32.8	4.30	10	−22
C12-DDAC	95	<MDL	0.139	0.0495	0.2	98	<MDL	2.91	0.047	0.1	−5
C14-DDAC	95	<MDL	0.050	0.0147	0.1	100	0.0002	0.462	0.016	0.04	9
C16-DDAC	100	0.0355	4.67	0.231	1.0	100	0.0031	4.24	0.374	0.9	62
C18-DDAC	100	0.0809	22.1	1.71	7.0	100	0.0192	33.1	3.47	7.9	103*
∑DDAC	100	1.35	41.4	8.87	30	100	0.0595	68.9	12.3	26	39
ATMACs											
C8-ATMAC	100	0.0007	0.253	0.0223	0.1	95	<MDL	0.507	0.057	0.1	156*
C10-ATMAC	100	0.0146	2.41	0.196	0.8	93	<MDL	6.76	0.266	0.6	36
C12-ATMAC	100	0.0166	22.5	0.758	3.2	100	0.0281	13.1	1.25	2.9	65
C14-ATMAC	95	<MDL	4.05	0.131	0.5	100	0.0034	2.51	0.275	0.6	110*
C16-ATMAC	100	0.246	14.0	2.20	9.3	100	0.0116	61.3	4.59	10	109*
C18-ATMAC	100	0.030	6.32	0.546	2.3	100	0.0096	9.80	0.841	1.9	54
∑ATMAC	100	0.698	26.1	6.36	22	100	0.235	66.5	8.78	18	38
∑QAC	100	6.55	127	36.3	100	100	1.95	531	58.9	100	62*

[a]MDL is the method detection limit. Asterisks indicate a statistical difference at the $p < 0.05$ level based on a Mann−Whitney test.

dust has long been recognized as a reservoir and a major human exposure pathway for many environmental contaminants, especially for children.[26,27] Due to their low volatility, QACs are easily adsorbed to solid airborne particles and dust, where they are unlikely to degrade. This leads to long-term contamination of the indoor environment, which is likely to last long after the pandemic.[28] Therefore, a better understanding of the increased exposure to QACs during and after the COVID-19 pandemic is essential for assessing its potential effects on human health.

This is the first study to investigate the occurrence of 19 QACs in residential dust collected before and during the outbreak of COVID-19. In addition, we also measured the levels of QACs in selected disinfecting products commonly used in sampled homes and evaluated the effects of using certain products and disinfection frequency on the levels of QACs in the indoor environment.

■ MATERIALS AND METHODS

Sample Collection and Analysis. Forty dust samples were collected from residential homes in Indiana in June 2020 (during the COVID-19 crisis in the United States). In addition, 21 dust samples collected from Indiana homes in 2018 and 2019 (before the COVID-19 outbreak) were obtained from the archives of the citizen-science program MapMyEnvironment. For both sample groups, dust from vacuum containers and bags (containing dust collected from the entire home) was transferred by the homeowner to resealable bags, delivered or shipped to the laboratory, and stored at room temperature until analysis. Information about the change in disinfecting habits (if disinfecting more frequently since the COVID-19 outbreak), the disinfection frequency (how many times per month or week), and commonly used cleaning products in sampled homes was also collected at the time of sampling during the pandemic. Cleaning products (sprays and wipes) indicated as frequently used in homes sampled during the pandemic were purchased from local markets for analysis.

All dust samples were sieved using a 500 μm mesh size sieve, and approximately 100 mg of dust was transferred to a glass tube, spiked with a surrogate standard (d_7-C12-BAC), sonicated in 4 mL of acetonitrile for 1 h, and centrifuged at 3000 rpm for 5 min. The supernatant was transferred into a clean tube, and the residues were re-extracted twice with 4 mL of acetonitrile. The combined extracts were concentrated to 1 mL using nitrogen gas and spiked with an internal standard (d_7-C14-BAC) used for quantification of the target analytes. For the analysis of disinfecting products, 10 μL of a product was diluted with 9.99 mL of acetonitrile, and then 1 mL of the diluted solution was spiked with the internal standard (d_7-C14-BAC) before the direct instrumental analysis.

The samples were analyzed using ultra performance liquid chromatography-triple-quadrupole mass spectrometry. The complete analyte list, details of the instrumental analysis, quality control and assurance measures, and data analysis are provided in the Supporting Information.

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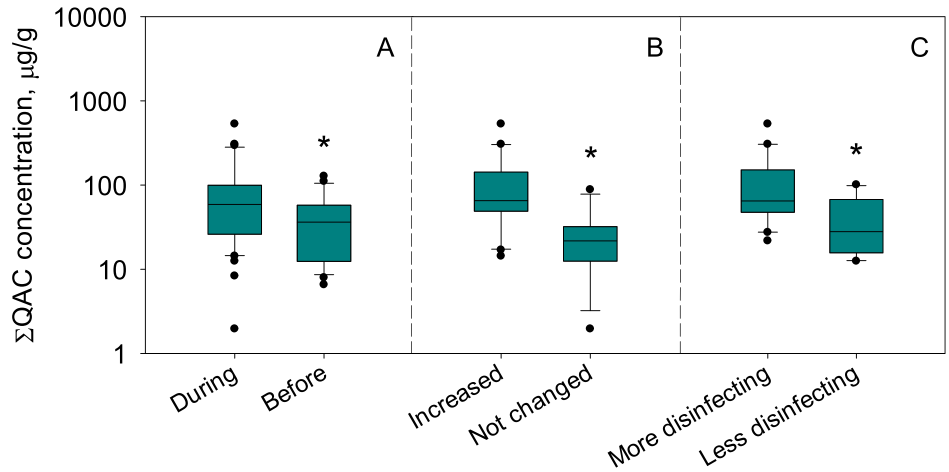

Figure 1. ∑QAC concentrations in dust collected from homes: (A) during (n = 40) and before (n = 21) the COVID-19 pandemic, (B) with increased (n = 29) and not changed (n = 11) disinfecting frequency during the COVID-19 pandemic, and (C) with more frequent (1−5 times per week; n = 27) and less frequent (less than once a week or without disinfecting chemicals; n = 13; three outliers were omitted) disinfecting. Concentrations are shown as box plots, representing the 25th and 75th percentiles; black lines represent the median, and whiskers represent the 10th and 90th percentiles. The asterisks indicate a statistical difference at the p < 0.05 level based on a Mann−Whitney test.

RESULTS AND DISCUSSION

Dust Concentrations. Each of the 19 QACs was detected in >90% of the samples collected during the COVID-19 pandemic (Table 1). The total QAC concentration (∑QAC, the sum of 19 QACs) ranged from 1.95 to 531 μg/g (median of 58.9 μg/g). Benzylalkyldimethylammonium compounds (BACs) made up the major group of QACs found in dust at a median ∑BAC concentration (the sum of seven BACs) of 27.1 μg/g. Dialkyldimethylammonium compounds (DDACs) and alkyltrimethylammonium compounds (ATMACs) were found at lower concentrations [median ∑DDAC (the sum of six DDACs) of 12.3 μg/g and ∑ATMAC (the sum of six ATMACs) of 8.78 μg/g]. BACs, DDACs, and ATMACs accounted for 56%, 26%, and 18% of the ∑QAC concentrations, respectively. C12- and C14-BACs were the most abundant QACs and contributed 29% and 22% to the ∑QAC concentrations, respectively. Among the DDACs and ATMACs, C10- and C18-DDACs and C16-ATMAC were the most abundant, respectively, and contributed up to 10% of the ∑QAC concentrations. Overall, these five compounds comprised ∼80% of the ∑QAC concentrations. This high proportion is likely related to large production volumes and to the wide application of these individual QACs.[15] Significant correlations were found among all QAC concentrations (Table S1), suggesting a common source for these compounds.

Similarly, all QACs were detected in >95% of the samples collected before the COVID-19 outbreak (Table 1), but at significantly lower concentrations than in samples collected during the pandemic [median of 36.3 μg/g; p < 0.05 (Figure 1A)]. Overall, the dust concentrations of 10 QACs have significantly increased during the pandemic compared to those of the dust collected before the pandemic (Table 1). The median ∑QAC concentration in samples collected during the pandemic increased by 62% when compared to the samples collected before the pandemic, with the largest increase of 91% found for BACs. Interestingly, the contributions of BACs, DDACs, and ATMACs to the ∑QAC concentrations (48%, 30%, and 22%, respectively) in prepandemic samples were similar to those found in dust collected during the pandemic, suggesting a similar source of QACs in both sample groups. These results indicate that the levels of QACs in the indoor environment have increased since the outbreak of COVID-19.

When compared with the levels of other environmental contaminants reported in dust from the United States, the median QAC concentration in this study was ∼3 times higher than that for organophosphate esters (16.8 μg/g)[26] and ∼1000 times higher than that for per- and polyfluoroalkyl substances (84.5 ng/g).[27] On the contrary, these QAC levels were ∼6 times lower than those for phthalates (median of 396 μg/g).[29] Incidentally, QACs were detected in urban estuarine sediment from New York state (median of 29 μg/g)[19] and in surface sediment from the Great Lakes (2.4−4.9 μg/g), but at concentrations lower than those measured here.[20] These lower environmental levels may be due to the effectiveness of the removal processes at wastewater treatment plants.[5]

Concentrations in Cleaning Products. Table S2 shows the QAC concentrations in seven cleaning and disinfecting products indicated as commonly used in the homes that were sampled during the pandemic. All three QAC groups were detected in the analyzed products, but at widely varying concentrations. Products 1 and 2 had the highest ∑QAC levels, reaching 16600 and 1350 mg/L and accounting for 1.66% and 0.135% by weight, respectively (similar to those indicated on the products' labels, 1.2% and 0.18% by weight, respectively). These concentrations were 10−1000 times higher than those in the rest of the products (2.52−156 mg/L). BACs were the predominant compounds in products 1−3, contributing 83%, 99%, and 98% to the ∑QAC concentrations, respectively (Figure S2). This contribution decreased to 0.4−23% in products 4−7. It should be noted that products 1 and 2 are included in the EPA's List N of disinfectants effective for SARS-CoV-2.[30]

Effect of Disinfecting Practices. Seventy-two percent of participants in this study indicated that they have increased the frequency of disinfecting in their homes since the beginning of the COVID-19 pandemic. Overall, the ∑QAC concentrations in homes with the increased disinfecting frequencies during the COVID-19 crisis (median of 65.2 μg/g) were significantly higher than in homes that did not change their disinfecting routine (median of 21.7 μg/g; p < 0.05) (Figure 1B and Table S3), suggesting that the intensified disinfecting practices can significantly increase exposure to QACs in the indoor environment.

The ∑QAC levels in homes that reported cleaning and disinfecting from one to a few times a week were significantly higher than in homes that did not do weekly disinfecting or use disinfecting chemicals [$p < 0.05$ (Figure 1C and Table S3)]. Overall, the homes with higher cleaning frequencies (1−5 times per week) had a ∑QAC concentration twice as high as that of homes with less frequent (<1 per week or without disinfecting products) cleaning (medians of 64.6 μg/g vs 28.0 μg/g; $p < 0.05$). A linear regression between the average ∑QAC concentrations of the 10 QACs for which the levels have significantly increased since the COVID-19 outbreak (see Table 1) and the disinfecting frequency in homes was highly significant [$r^2 = 0.9933$; $p = 00034$ (Figure S3)], further indicating that the disinfecting practices can have a strong effect on the indoor QAC levels.

Ninety percent of households reported using a disinfecting product for their cleaning routine, and >80% of these households regularly used products 1, 2, and 7. Figure 2 compares the average contributions of BACs, DDAC, and ATMACs in these three products and in dust samples from the homes that regularly used only these three products. These contributions in dust were similar to those in products (58%, 24%, and 18% vs 64%, 14%, and 22%, respectively). The similarity between the profiles in dust and products suggests that disinfecting products frequently used in homes could be a significant source of these compounds.

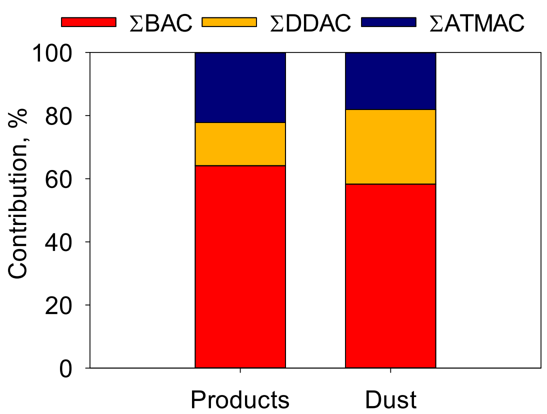

Figure 2. Comparison of the average contributions of the three QAC groups to the ∑QAC concentrations in house dust and in the only three disinfecting products (products 1, 2, and 7) used in >80% of the homes.

Exposure Assessment. The estimated daily intakes (EDIs) of QACs during the COVID-19 pandemic via dust ingestion were calculated for toddlers and adults for the homes with higher disinfecting frequencies (1−5 times per week) and for the homes with less frequent cleaning (less than once a week) (Table 2). The highest ∑QAC EDI [615 ng/kg body weight/day] was found for toddlers in homes with higher disinfecting frequencies and was up to 10 times higher than that estimated for adults. The EDIs for BACs and DDACs were below the tolerable daily intake thresholds for these two compound groups [1×10^5 ng (kg of body weight)$^{-1}$ day^{-1}] established by the European Food Safety Authority (EFSA).[31]

Limitations and Implications. This study had several limitations. The sample size was small for both dust and products due to the efforts to finish the study during the time period of the COVID-19 pandemic, and the samples were collected from a limited geographic area. Dust was collected by citizen scientists from the vacuum bags and canisters due to the inability to enter the homes during the pandemic, and it was not possible to determine which rooms were sampled. The samples collected before the COVID-19 pandemic were collected from homes different from the homes from which samples were collected during the pandemic.

Table 2. Estimated Daily Intakes (EDIs, ng/kg bw/day) of Each QAC Group via Dust Ingestion for Toddlers and Adults in Homes with More Frequent (1−5 times per week) and Less Frequent (less than once a week or without disinfecting chemicals) Disinfecting during the COVID-19 Pandemic

	more disinfecting		less disinfecting	
	toddlers	adults	toddlers	adults
∑BAC	423	36.3	94.9	8.13
∑DDAC	106	9.07	64.8	5.55
∑ATMAC	86.7	7.43	46.2	3.96
∑QAC	615	52.7	206	17.6

Nonetheless, this is the first study to assess human exposure to a wide suite of QACs in the indoor environment. The timing of this study is important considering the increased use of disinfectants due to the current COVID-19 pandemic. Our findings indicate that the indoor exposure to QACs is widespread and significantly higher in households with increased disinfecting frequencies due to the pandemic. The similarity between the profiles of QACs in products and dust collected from the same households suggests that the disinfecting products are a significant source of these compounds in homes. As the COVID-19 pandemic continues, the use of these compounds is expected to increase worldwide, and more research is needed to confirm our findings in other locations. Furthermore, more intense disinfecting procedures are advised for care facilities, schools, and other high-risk places, many of which serve populations most vulnerable to these exposures. Our findings call for urgent research on risks associated with the increased exposure to these chemicals.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.0c00587.

Information about chemicals used in this study, instrumental methods, and quality control and assurance measures; correlations among QAC concentrations in dust; dust QAC concentrations grouped on the basis of the disinfecting frequency; QAC concentrations and patterns in cleaning products; and correlation between the disinfecting frequency and QAC dust concentrations (PDF)

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Notes

The authors declare no competing financial interest.

A version of this paper prior to peer review is available on preprint servers at Indiana University[32] and ChemRxiv.[33]

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