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Volatile organic compounds emitted by conventional and "green" cleaning products in the U.S. market

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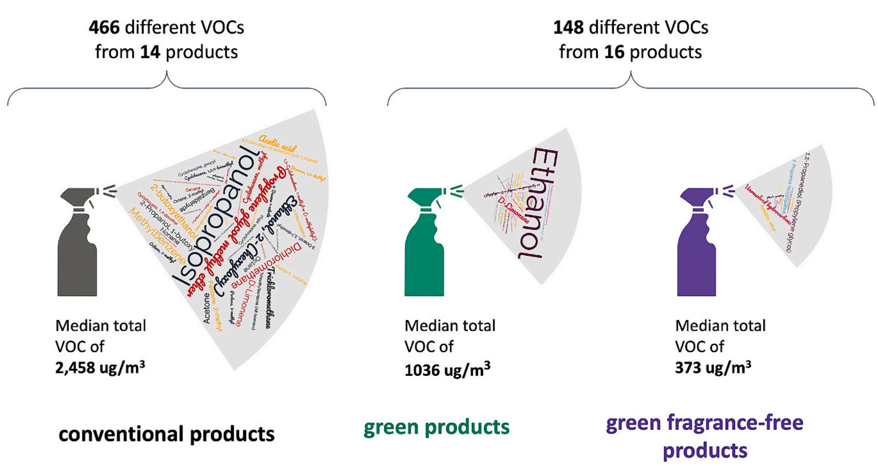
HIGHLIGHTS

- Air chamber testing of 28 cleaning products and 2 air fresheners detected 530 VOCs.
- The number and concentrations of VOCs were lower in "green" products.
- Fragrance-free products had significantly lower numbers and concentrations of VOCs.
- Exposure limits were used to calculate relative risk for each product.

GRAPHICAL ABSTRACT



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ABSTRACT

Exposure to cleaning products has been associated with harm to the respiratory system, neurotoxicity, harm to the reproductive system, and elevated risk of cancer, with greatest adverse impacts for workers exposed in an occupational setting. Social and consumer interest in cleaning products that are safer for health created a market category of "green" products defined here as products advertised as healthier, non-toxic, or free from harmful chemicals as well as products with a third-party certification for safety or environmental features. In the present study we examined the air quality impacts of cleaning products and air fresheners, measuring the number, concentrations, and emission factors of volatile organic compounds (VOCs) in an air chamber following product application. Across seven common product categories, 30 products were tested overall including 14

Abbreviations: IFRA, International Fragrance Association; VOC, Volatile Organic Compound; C&L Inventory, Classification and Labeling Inventory; ECHA, European Chemicals Agency; DTSC, California's Department of Toxic Substances Control.

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conventional, 9 identified as “green” with fragrance, and 7 identified as “green” and fragrance-free. A total of 530 unique VOCs were quantified with 205 additional VOCs detected below the limits of quantification. Of the quantifiable VOCs, 193 were considered hazardous according to either the California’s Department of Toxic Substances Control Candidate Chemicals List or the European Chemical Agency’s Classification and Labeling Inventory. The total concentration of VOCs and total emission factors across all products with detections ranged from below limits of detection to 18,708 μg/m^3, 38,035 μg/g product and 3803 μg/application. Greater total concentration, total emission factors, and numbers of VOCs were generally observed in conventional cleaning products compared to products identified as “green”, particularly compared to fragrance-free products. A hazard index approach was utilized to assess relative risk from measured VOC emissions. The five products with the highest hazard indices were conventional products with emissions of 2-butoxyethanol, isopropanol, toluene and chloroform. Overall, this analysis suggests that the use of “green” cleaning products, especially fragrance-free products, may reduce exposure to VOC emissions.

1. Introduction

Cleaning product application in both occupational and home settings is associated with harm to the respiratory system and other chronic health problems (Dumas and Le Moual, 2020). The use of cleaning products results in inhalation exposure to numerous volatile organic compounds (VOCs) and particulates (Vu et al., 2017). The greatest risk of adverse long term health outcomes due to cleaning products is observed in highly exposed individuals, particularly people who do professional cleaning in commercial and residential settings.

Epidemiological research established a clear association between cleaning and harm to the lungs and respiratory system (Van den Borre and Deboosere, 2018). In a large cohort study of women in Europe, a decline in lung function was observed in women who cleaned at home as well as professionally, when using cleaning sprays as well as other cleaning agents (Svanes et al., 2018). A meta-analysis found that occupational exposure to cleaning products was associated with a 50 percent increased risk for asthma and a 43 percent increased risk of chronic obstructive pulmonary disease (Archangelidi et al., 2020). An increased risk of lung cancer has been observed in women employed as cleaners (Atramont et al., 2016), as well as increased risk of respiratory and cardiovascular mortality (Van den Borre and Deboosere, 2018).

Exposure to cleaning products also harms children’s health. Frequent household use of cleaning products during infancy, especially use of scented spray products, was associated with a higher risk of childhood asthma and wheezing (Bably et al., 2021; Parks et al., 2020). Similarly, exposure to cleaning products daily during infancy or pregnancy was associated with persistent wheezing in early childhood and increased lower respiratory tract infections (Casas et al., 2013; Herr et al., 2012; Sherriff et al., 2005). A recent investigation found that exposure to cleaning products during pregnancy resulted in a greater risk for children developing asthma (Tjalvin et al., 2022). However, a cross sectional analysis of asthma in adolescents and frequency of use of cleaning products observed no association (Bukalasa et al., 2019).

VOCs commonly emitted from cleaning and other consumer products have been investigated as the cause for respiratory harms and have been associated with other chemical hazards including cancer, neurotoxicity, and reproductive toxicity. Direct measures of VOC emissions from cleaning and consumer products detects hundreds of compounds, the vast majority of which do not appear on ingredient labels or safety data sheets (Nematollahi et al., 2018; Steinemann, 2009; Steinemann et al., 2011). Fragranced products in particular have been implemented in causing self-reported health problems in nearly 35% of the general population in the United States (Steinemann, 2016).

In addition to VOC emissions from cleaning products as one source of VOCs in indoor air, there are numerous other VOC sources including cooking, use of heating appliances, air fresheners, pesticides, consumer products, furniture and building materials (Paciencia et al., 2016; U.S. EPA, 2022; Zhou et al., 2022). Better characterization of VOC emissions in terms of chemical prevalence, concentration, and associated health hazards is needed to understand air quality and respiratory health impacts of indoor use of cleaners (Clausen et al., 2020). Overall, indoor air has higher VOC levels than outdoor air (Paciencia et al., 2016; Xu et al., 2016) and VOC emissions from consumer products can negatively impact outdoor air quality (McDonald et al., 2018). There is ongoing research on characterizing the human health impacts of indoor VOC exposure (Zhou et al., 2023) such as adverse pulmonary effects (Alford & Kumar, 2021).

Greater public awareness regarding indoor air quality and consumer interest in cleaning products that are safer for health prompted marketplace introduction of products advertised as “green” or fragrance-free as well as products carrying a third-party certification for safety or environmental features. Published literature comparing this new “green” market sector to “conventional” cleaning products remains limited. A California-based investigation of VOC exposure from cleaning products found that switching from conventional to green cleaning products resulted in a significant reduction in the air concentration of several VOCs associated with health hazards (Harley et al., 2021). Subsequent laboratory testing found that the highest VOC emissions originated most often from conventional products (Calderon et al., 2022). Similar results were reported in a study conducted in Massachusetts (Lindberg et al., 2021). In contrast, several articles published by a research group in Australia found no difference between conventional and green products (Nematollahi et al., 2018, 2019; Steinemann et al., 2021). Here, we investigated VOC emissions from a group of 30 products purchased in the United States, comparing overall VOC emissions and the presence of potentially hazardous VOCs in conventional, green and fragrance-free cleaning products.

2. Methods

2.1. Criteria for product selection

Thirty products used for a variety of cleaning functions were purchased from online retailers between December 2019 and May 2022 and shipped directly to the testing lab (Fig. 1). Within this group, 16 were classified as conventional, 14 were green of which 9 were classified as green with fragrance and 7 were classified as both green and fragrance-free. Product categories included air freshener and a variety of cleaners, including all-purpose, carpet, floor, glass, and wood cleaners as well as laundry stain removers. Product selection was additionally guided by prioritizing popularity as evaluated by both availability and the number of customer reviewers on national retailer websites including Walmart, Home Depot, and Amazon. “Green” products were identified based on product marketing claims stating the product is safer, healthier, non-toxic, free from harmful chemicals as well as by the presence of a third-party certification from U.S. EPA Safer Choice, Green Seal or UL ECOLOGO. “Fragrance-free” products were identified according to manufacturer statements on product packaging. “Conventional” products were identified in 2 ways: (1) by the absence of “green” claims or certifications; and (2) by the disclosure of ingredients of concern according to chemical listings published by authoritative U.S.-based and international agencies and organizations. These ingredients of concern identified in conventional products we tested are listed in Supporting

Information Table S1, and include both volatile ingredients such as 2-butoxyethanol and non-volatile ingredients such as dyes. Products were selected to include green and conventional products for each category as well as for different product forms. Each product selected for testing represented a unique brand. The ingredient information disclosed on the company websites and listed in Supporting Information Table S1 was compiled after the air sampling was completed and could potentially represent a distinct formulation under the same product name. Product reformulation and/or changes in product names may have occurred since the time period when products were purchase for testing.

2.2. Air chamber testing methodology

Air chamber testing was performed between January and July 2022 by a leading U.S.-based testing organization in their indoor air quality laboratory accredited to the requirements of ISO 17025, "Testing and calibration laboratories" (www.iso.org/standard/66912.html). The testing organization has a long and consolidated expertise in product emissions testing. All products except air fresheners were tested using a modified ASTM D 5116, "Standard Guide for Small-Scale Environmental Chamber Determinations of Organic Emissions from Indoor Materials/Products" ASTM, West Conshohocken, PA, (www.astm.org/d5116-17.html) method in a controlled dynamic environmental chamber supplied with purified air to minimize background contamination. Two air fresheners were tested according to ASTM Standard D6670-01, "Standard Practice for Full-Scale Chamber Determination of VOCs from Indoor Materials/Products" (www.astm.org/d6670-01.html).

All products were shipped to the testing laboratory unopened and unblinded where they were stored in a controlled environment until testing occurred. An air chamber size of 0.0945 m^3 was used for most products, except air fresheners which were tested in a whole room chamber measuring 31 m^3. Air chambers were constructed of high-polished stainless steel with inlet and outlet air manifolds for uniform air mixing. The ventilation rate in both chambers was 1±0.05 change per hour. Tests were conducted at a temperature of 23 °C ± 3° and a relative humidity of 50% ± 5%. Between tests, chambers were opened and flushed with multiple air changes. Background levels were taken from empty chambers at least one day prior to testing to verify levels below <10 μg/m^3 total VOCs, <10 μg/m^3 total particles, <2 μg/m^3, formaldehyde, <2 μg/m^3 for any individual VOC. For the non-air freshener products, a blank glass slide (0.0516 m^2) and a clean microfiber cloth were acclimated for at least 4 h in the empty chamber before taking a product-loaded chamber measurement. The unopened product or pre-weighed amount of product in a sealed container was then added to the chamber and left either overnight or 24 h. The following day the product was opened and applied to the glass slide in an amount representative of use and, if applicable, was wiped off at least 1 min later, with the cloth remaining in the chamber for the duration of testing. Product application rates were guided by manufacturer's directions and previous research on chamber testing of cleaning products, which conducted a survey of real-world application rates for a few types of cleaning products and used about 11 g/m^2 to 17 g/m^2 for all purpose spray type products and liquid non-spray products respectively and 23–31 g/m^2 for floor products (Singer et al., 2006b). For all-purpose, degreaser, disinfectant, glass, laundry, and bathroom products sold in trigger spray bottles, generally one full spray was applied to the glass slide. Stain remover and carpet cleaner products received two full sprays to approximate the saturation required for the most effective cleaning method directed by manufacturers. Wipe and mopping cloth products utilized a single wipe and were applied until the surface became visibly wet. Products were generally applied as sold except three products were diluted with deionized water according to manufacturer directions. Air sampling occurred over 4 h, starting just before the sample was opened.

Fig. 1. Summary chart of 28 household cleaning products and 2 air fresheners tested for volatile organic compound (VOC) emissions during simulated usage. Products were categorized as either conventional (n = 16), green (n = 9) or green fragrance-free (n = 7) represented by grey, green and purple shading respectively. Products are grouped by cleaning purpose categories, with product form and number of products of each type indicated. Product names, packaging or product form, claims and certifications, and ingredient information as available are listed in Supporting Information Table S1. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

The hand spray air freshener was sprayed 5 times at minute 0 and 5 times at minute 90 with VOC emissions collected for 4 h. The automatic air freshener was set to spray every 9 min with VOC emissions also collected for 4 h. Supporting Information Table S2 provides experimental details for each product tested including the product category, chamber background preparation, loading time, production application details, including usage in grams and elapsed time before wiping the product. The amount of each product applied varied due to differences in manufacturing components, product density (solid vs. liquid), the presence of a substrate such as wipes, and product use characteristics.

VOC emissions were analyzed using a sorbent collection and thermal desorption into the GC/MS with methodology adapted from ISO 16000-6, "Indoor air – Part 6: Determination of volatile organic compounds in indoor and test chamber air by active sampling on Tenax TA sorbent, thermal desorption and gas chromatography using MS or MS-FID" (www.iso.org/standard/73522.html), U.S. Environmental Protection Agency "Compendium of Methods for the Determination of Toxic Organic Compounds in Ambient Air Second Edition Compendium Method TO-17 Determination of Volatile Organic Compounds in Ambient Air Using Active Sampling Onto Sorbent Tubes. EPA/625/R-96/010b" (www.epa.gov/amtic/compendium-methods-determination-toxic-organic-compounds-ambient-air) and ASTM D 6196, "Practice for the Selection of Sorbents and Pumped Sampling/Thermal Desorption Analysis Procedures for Volatile Organic Compounds in Air" (https://www.astm.org/d6196-15e01.html). VOC samples were collected using mass flow controllers at a rate of 0.05 L/min for 240 min and aldehydes were sampled at 0.1 L/min over the same time. Samples were collected through Tenax TA and Tenax TA with Carbosieve SIII for VOCs and DNPH cartridges for aldehydes. VOCs were measured by GC/MS according to ISO 16000-6 and aldehydes were measured by HPLC according to ISO 16000-3 "Indoor air – Part 3: Determination of formaldehyde and other carbonyl compounds in indoor air and test chamber air – Active sampling method" (www.iso.org/standard/81864.html). Individual chemicals results from the ISO 16000-3 testing methodology were only used for VOCs with 4 or fewer carbon atoms: 2-butenal (4170-30-3), acetaldehyde (75-07-0), butanal (123-72-8), formaldehyde (50-00-0), propanal (123-38-6) (Yrieix et al., 2010). VOC total concentration was calibrated relative to toluene equivalence with individual VOC concentrations based on multipoint calibration standards as indicated in the full table of results, or relative to toluene when a standard was not available.

Quality control procedures within the laboratory testing environment included weekly supply air purity monitoring, multi-point calibration and linear regression of all standards, control analysis for chamber background, and instrumentation. The chamber operation and control were completed to meet the standards of ASTM D 5116 and ISO 16000-9, "Indoor air —Part 9: Determination of the emission of volatile organic compounds from building products and furnishing — Emission test chamber method" (www.iso.org/standard/38203.html).

2.3. VOC identification and quantification

A library of compound retention time and mass spectra was used first to identify matches for individual VOCs. Additional substance identification was completed using the U.S. National Institute of Standards and Technology general mass spectral library. Loaded chamber background levels were subtracted from VOC concentrations used in subsequent analyses. When the loaded chamber background level for a VOC was not detected, the individual VOC limit of detection (2 $\mu g/m^3$) was subtracted from the measured value. Total VOC concentration for each product was calculated by adding together the concentrations emitted for all quantifiable VOCs. VOC emissions were also calculated as emission factors, expressed as μg/application unit such as spray, wipe or pour, and μg/g product using the grams emitted from the product and the ventilation rate in the chamber (CDPH, 2010) For comparisons of VOC concentrations and emission factors we examined cleaning products (all-purpose, carpet, floor, glass, wood cleaners and stain removers) separately from air fresheners since air fresheners were tested in larger chambers and have different use patterns compared to other products in our study. For comparing numbers of VOCs emitted per product we considered all product types.

2.4. Identification of hazardous VOCs

For the purposes of this analysis, VOCs were classified as hazardous based on CAS number matching to two authoritative chemical hazard lists, California's Department of Toxic Substances Control (DTSC) Candidate Chemicals List and the European Chemicals Agency (ECHA) Classification and Labeling Inventory (C&L Inventory). California DTSC maintains a list of hazardous chemicals known as the Candidate Chemicals List (https://dtsc.ca.gov/scp/candidate-chemicals-list/; accessed November 2021). According to the DTSC definition, a candidate chemical is a chemical that exhibits a "hazard trait and/or an environmental or toxicological endpoint" identified as such based on the State of California regulations. Chemicals on the list are associated with neurotoxicity, carcinogenicity, developmental and reproductive toxicity, endocrine disruption, target organ toxicity (liver, urinary system, eyes, digestive system) as well as chemicals that are persistent, bioaccumulative and toxic.

Additionally, under the European Union's Classification, Labeling and Packaging Regulation, physical, environmental and human health hazards associated with chemicals are reported to the ECHA by manufacturers and importers, referred to as self-classification, and this information is maintained in the C&L Inventory (https://echa.europa.eu/information-on-chemicals/cl-inventory-database). The database was downloaded in November 2021, including chemical information for 195,769 chemicals. Of those, 189,934 chemicals had hazard information. For the present study, we considered a subset of human health hazards listed in the C&L Inventory including skin sensitization, respiratory sensitization and irritation, carcinogenicity and genotoxicity, developmental and reproductive toxicity, and acute inhalation toxicity. Further, our analysis included both harmonized and non-harmonized hazard classifications, whereby "harmonized" refers to classifications reviewed by the ECHA Risk Assessment Committee which have consistent hazard labeling throughout the European Union, while "non-harmonized" refers to self-reported or self-classified hazard classifications by manufacturers or importers of chemicals. A full list of hazard codes and statements used to identify VOCs hazardous to human health is listed in Supporting Information Table S3. The table of all VOCs identified above detection limits and their health hazards according to the DTSC Candidate Chemicals List and ECHA C&L Inventory hazards are shown in Supporting Information Table S4.

Detected VOCs were also matched by CAS numbers to several regulatory and industry databases including the U.S. EPA Toxic Substances Control Act Inventory (https://www.epa.gov/tsca-inventory/how-access-tsca-inventory), the complete ECHA C&L Inventory, the International Fragrance Association Transparency List (https://ifrafragrance.org/priorities/ingredients/ifra-transparency-list), and the Cleaning Product Ingredient Safety Initiative database published by the American Cleaning Institute, a manufacturers' association (https://www.cleaninginstitute.org/industry-priorities/science/cleaning-product-ingredient-safety-initiative-cpisi).

2.5. Cumulative risk assessment of VOCs emitted from cleaning products

To assess the potential health risks from exposure to the VOCs present in the products tested, we utilized a Hazard Quotient and Hazard Index approach, comparing the emitted concentrations for each VOC from a product to exposure limit values from government agencies. We identified VOCs for which exposure limits existed using a tiered approach. First, we identified 15 chemicals for which a Reference Exposure Level was published by the State of California Office of Environmental Health Hazard Assessment (https://oehha.ca.gov/air/genera l-info/oehha-acute-8-hour-and-chronic-reference-exposure-level-rel-summary). Next, an additional set of reference values for 32 chemicals was identified from the U.S. Occupational Safety and Health Administration (OSHA) database, https://www.osha.gov/annotated-pels. Lastly, for the 100 chemicals that contributed the most to total VOC emissions, additional information about exposure limits set by authorities in countries other than the United States was saught from the GESTIS Substance Database published by the German Social Accident Insurance and accessible at https://gestis-database.dguv.de/. For the exposure limits compiled from the GESTIS database, we selected the lowest 8-h limit value when available. In total, we included exposure limits for 67 VOCs detected during product applications, representing 48% percent of the total VOC emissions from all products. Supporting Information Table S4 provides a list of the exposure limits used. For each product, a hazard quotient was calculated for each VOC for which an exposure limit had been identified by dividing the quantified concentration by the exposure limit. A hazard index for each product was then calculated by adding together all hazard quotients for a given product.

2.6. Statistical analysis

Data were assessed for Gaussian distribution by D'Agostino & Pearson normality test and Shapiro-Wilk normality test. If the data were normally distributed, a one-way ANOVA with Tukey's multiple comparisons test was used to compare differences in concentration, emission factors, and number of VOCs, as well as calculated hazard quotients, between conventional and green fragrance or green fragrance-free products. If the variance between the groups was statistically significant using Brown-Forsythe test, the ANOVA was run using a Brown-Forsythe correction and Dunnett's multiple comparisons test. If the data were not normally distributed, a nonparametric test, the Kruskal-Wallis test and Dunn's multiple comparisons test, was used to assess differences. All statistical analyses were performed in Graph Pad Prism 9.

3. Results

3.1. VOC identification and quantification

Across all products tested, a total of 736 different VOCs were identified, with 530 compounds identified above the laboratory's limits of quantification (Fig. 2). The 14 conventional products accounted for 75.5% of the total VOC emission with the remaining 16 products contributing under 25% (21% green and 3.8% green fragrance-free), expressed as VOC chamber concentration. Not including ethanol, primarily emitted from a single product we classified as "green", 88% of VOC emissions were from conventional products, 8% from green with fragrance and 4% from green fragrance-free. Ethanol and isopropanol were both detected in emissions from 5 products and accounted for 26% of the total concentration of VOCs emitted from all products. Of note, the majority of total emissions of ethanol and isopropanol were from single separate products, contributing 69% and 96% of emissions for ethanol and isopropanol, respectively. The next eight chemicals contributing to overall total emissions were propylene glycol ether, 2-butoxyethanol, propylene glycol, D-limonene, propylene glycol butyl ether, 2-hexoxyethanol, acetic acid, and methylene chloride. The top 15 VOCs accounted for 51% of emissions, and the top 50 accounted for 71% of emissions. All but one product (which was a green, fragrance-free all-purpose cleaner) had detectable levels of VOCs. The total concentration of VOCs across all products with detections ranged from 21 to 18,708 μg/m3 (Fig. 3A). Supporting information Table S5 provides the results for the product application testing and the background for each VOC.

We next conducted an analysis of VOC emission factors on a μg per gram product basis, and a μg per application (spray, pour, wipe etc.) scenario. With the exception of the product with no detectable VOCS, total VOC emission factors ranged from 0.97 to 38,035 μg/g (Fig. 3B) and 1.9–3803 μg/application (Fig. 3C). For the conventional air freshener VOCs emissions per gram of product corresponded to 38,035 μg/g, orders of magnitude higher than other products, while the amount of VOC's emitted per gram of the "green" air freshener was 2792 μg/g. In

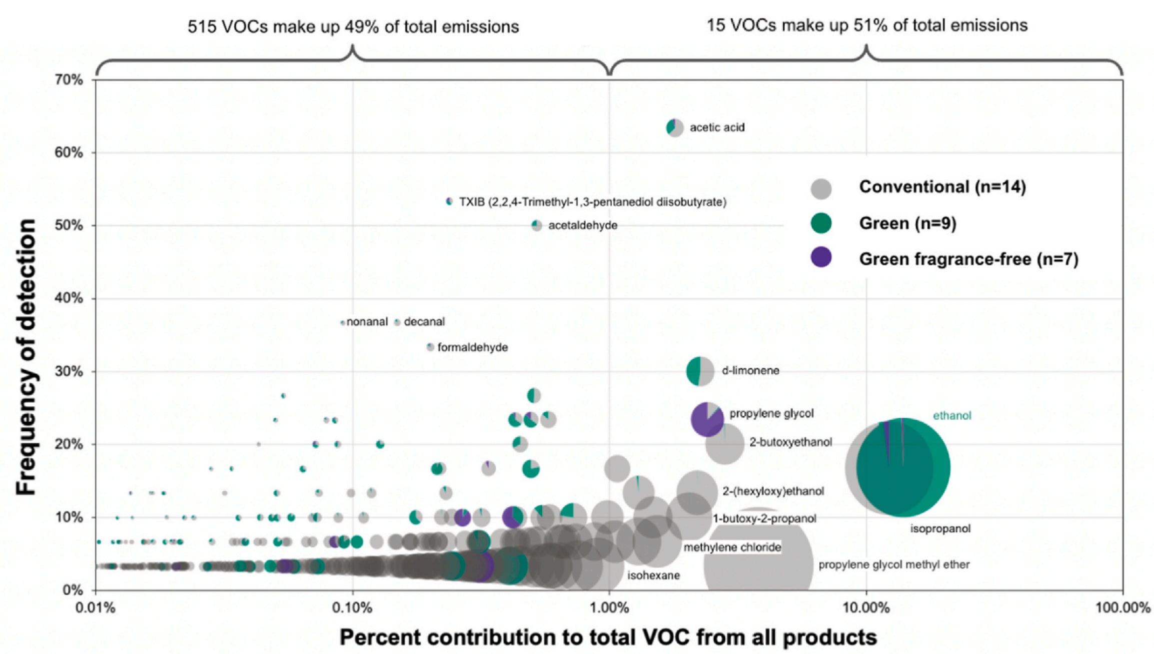

Fig. 2. Detection frequency and percent contribution to the total concentration of VOCs emitted for 530 detected VOCs from 30 products. Each detected VOC is represented by an individual circle representing the average concentration when detected. Shading within the circle represents the contribution from each product category (conventional, green, or green fragrance-free) to the sum concentration emitted for a given chemical. VOCs frequently detected (>30% of products) and those with the highest average concentration are labeled. Supplementary Figure S1 shows the same analysis when calculated on a μg/g basis for product emissions. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

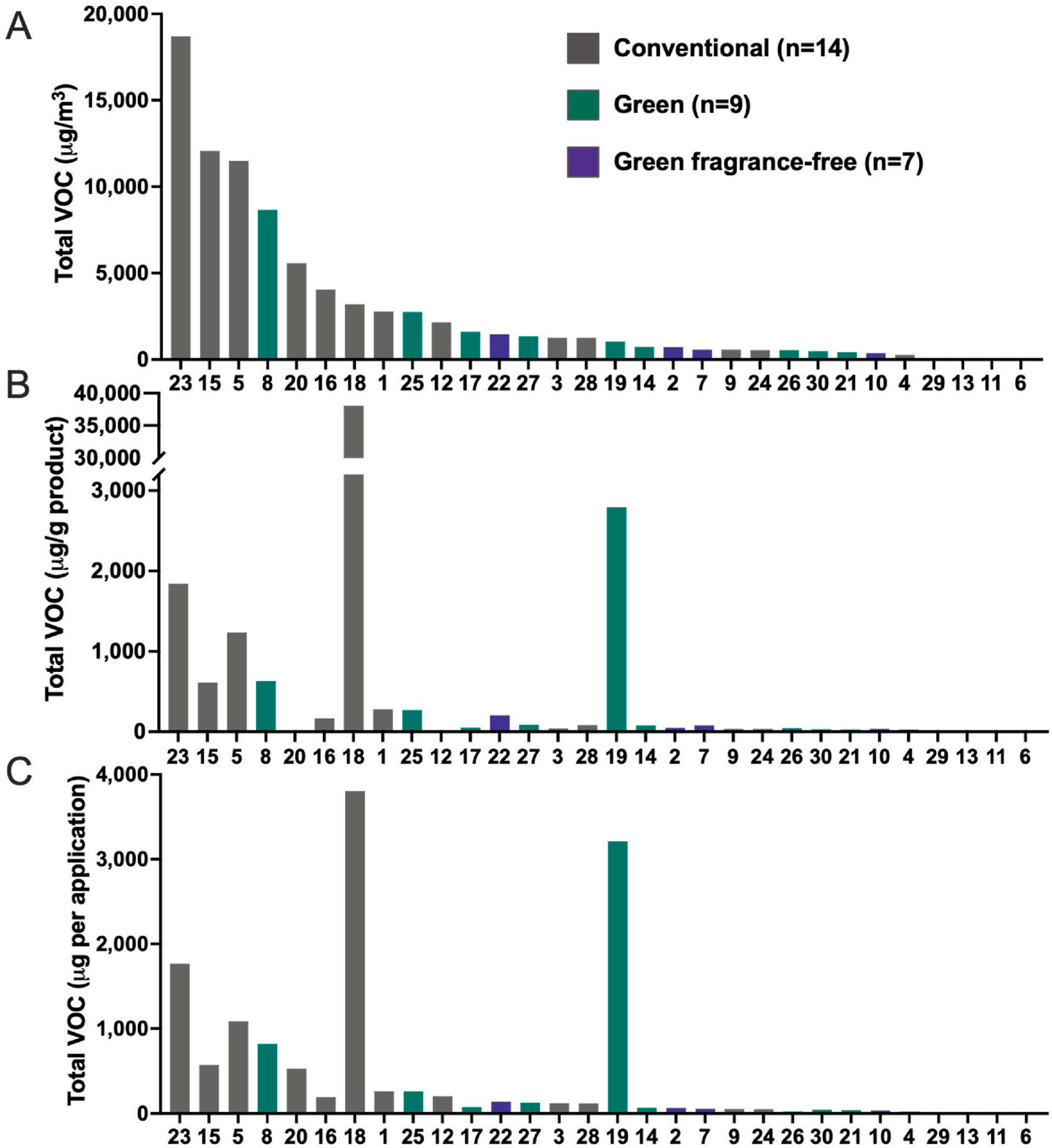

Fig. 3. Total VOC from individual products. Total VOCs are represented as A) concentration in μg/m3, B) μg per g product and C) μg per application. In Panel A, all 30 products are sorted based on VOC concentration (from high to low), and the same product order is used for panels B and C. Total VOC was calculated by adding together the value of each VOC emitted for a given product. Product IDs are listed in Supplemental Information. Of note, conventional air freshener is Product 18, while "green" air freshener is product 19. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

contrast, when assessing emissions on a μg per application (spray) scenario, the two air fresheners were comparable, emitting 3803 and 3210 μg per spray (Fig. 3).

For the group of 28 cleaning products (not including 2 air fresheners), calculations on a μg per gram product basis were comparable to total, non-normalized emissions described above. Specifically, 13 conventional products accounted for 73% of emissions and the remaining 15 green products accounting for 27% of emissions (20% green and 6% green fragrance-free). Ethanol and isopropanol remained the greatest contributors, accounting for 32% of emissions. The next eight chemicals, while the order shifted slightly and toluene replaced 2-propanol, 1-butoxy, were largely the same group and accounted for 53% of emissions, and the top 15 chemicals accounted for 58% of emissions (Figure S1).

Six VOCs were detected in 30% (9) or more of products: acetic acid, 2,2,4-trimethyl-1,3-pentanediol diisobutyrate (TXIB), acetaldehyde, nonanal, decanal, and formaldehyde. Of the 530 VOCs emitted from 30 products, 71% of VOCs were detected just once in emissions from individual products with 377 detected once, 81 VOCs detected twice, and 72 VOCs detected in the emissions from 3 or more products. Together, these results indicate a broad diversity of VOCs across all products as well as unique VOCs present in each product.

Of the 530 identified VOCs, 193 were considered hazardous, including 42 VOCs listed on the California DTSC Candidate Chemicals List and 191 VOCs present on the ECHA C&L Inventory, with 40 found on both lists (Table S4). The most common hazards identified for chemicals listed on the DTSC Candidate Chemicals List, were neurotoxicity (n = 14), carcinogenicity (n = 13), hepatotoxicity and digestive system toxicity (n = 13), respiratory toxicity (n = 11 and reproductive toxicity (n = 10). The most common hazards identified for chemicals with self-classifications on the ECHA C&L Inventory were respiratory toxicity such as irritation or acute toxicity for inhalation (n = 171), skin sensitization (n = 61), reproductive toxicity (n = 35), and carcinogenicity (n = 23).

3.2. Comparisons between conventional, green fragranced, and green fragrance-free products

When comparing the conventional to green air freshener, the conventional freshener resulted in three times greater concentration of VOCs, emitted four times as many VOCs and nearly 14 times the amount (μg) of VOCs per gram of product compared to the green air freshener, while grams per spray were similar between the two.

When comparing the concentrations of VOC emissions from conventional, green fragranced, and green fragrance-free products, both green fragranced and green fragrance-free had lower average concentrations (n = 28) and μg emitted per application (spray, wipe, pour; n = 28) of total and hazardous VOCs compared to conventional products, and the VOC emissions differences for green fragrance-free products reached statistical significance (Fig. 4A and B). A similar pattern was observed when considering the μg per gram product, although these findings were not statistically significant (Fig. 4C). The total number of VOCs per product ranged from 0 to 119, the number of hazardous VOCs on the ECHA C&L Inventory ranged from 0 to 40, and the number on the California DTSC Candidate Chemical list ranged from 0 to 16. Compared to conventional products, green fragrance-free products emitted a statistically significant lower number of total and hazardous VOCs on both lists, and green fragrance products emitted a statistically significant lower number of VOCs considered hazardous by California DTSC (Fig. 4D, n = 30). The VOC concentrations from products were not statistically significantly different across product categories or product forms (Figure S2), although some product forms yielded much higher product weights, and difference in grams emitted from similar product types was observed (Table S1), which could contribute to the observed differences in air chamber concentrations.

3.3. Hazard index analysis

Hazard indices were calculated for each product to estimate potential health risks associated with the mixture of VOCs emitted from products (Fig. 5). Although not statistically significant, green fragranced and green fragrance-free products had lower hazard indices than conventional products (Fig. 5A). The top five products with the highest hazard indices were conventional products. In the product with the

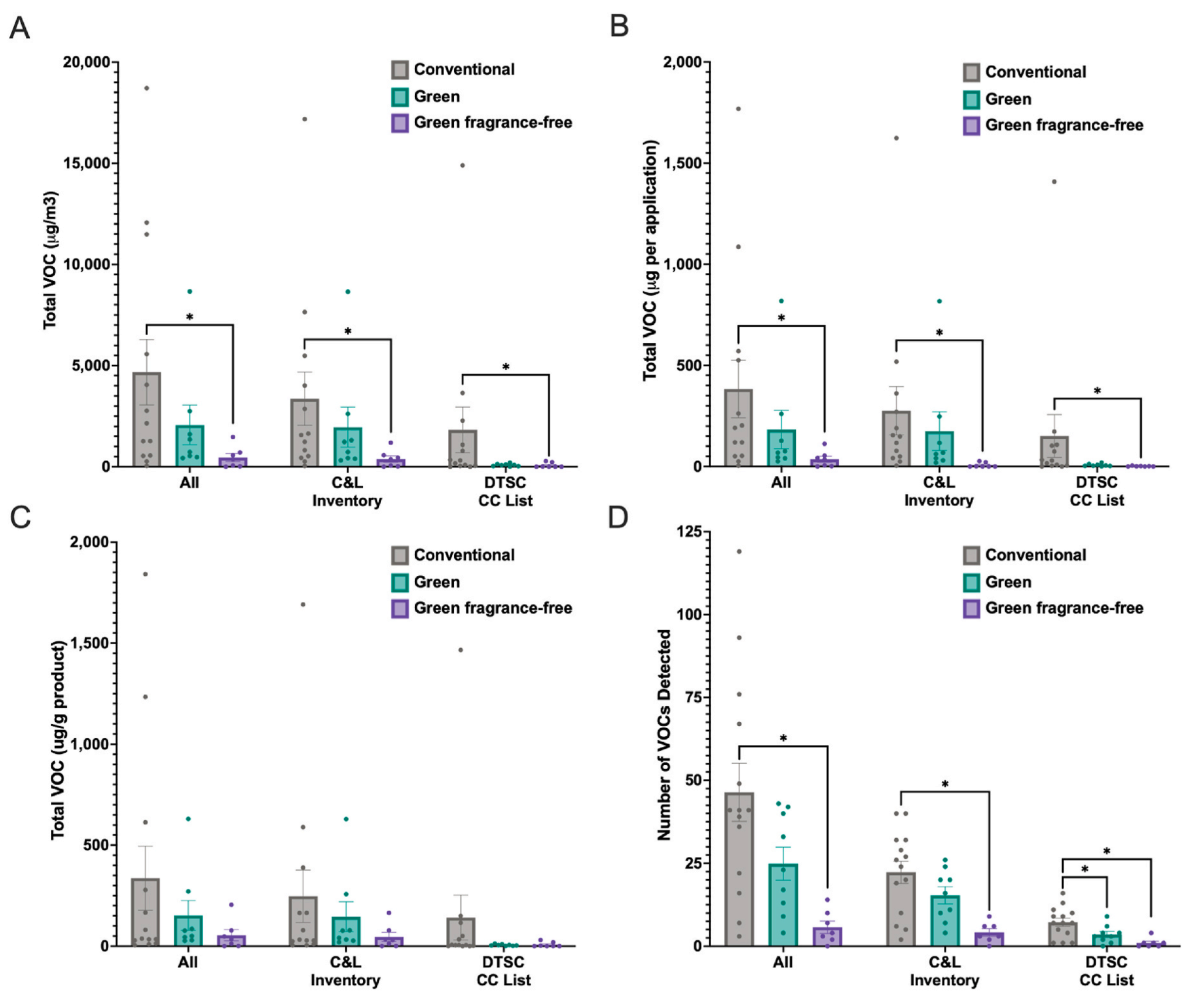

Fig. 4. Total and hazardous VOC emissions from conventional, green, and fragrance-free cleaning products. VOCs included in the analysis were all VOCs, hazardous VOCs identified on either the ECHA Classification & Labeling Inventory (C&L Inventory) or California's Department of Toxic Substances Control (DTSC) Candidate Chemicals List (DTSC CC List). The bars represent average values (± standard error) with individual products represented by dots. Panel A, total VOC concentration; n = 28 products. Panel B, μg VOCs per application; n = 28 products. Panel C, total VOC, μg per g product; n = 28 products. Panel D, number of VOCs; n = 30 products. Statistical significance between groups based on one-way ANOVA with Tukey's post-hoc test is indicated by the asterisk symbol (*; $p < 0.05$). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

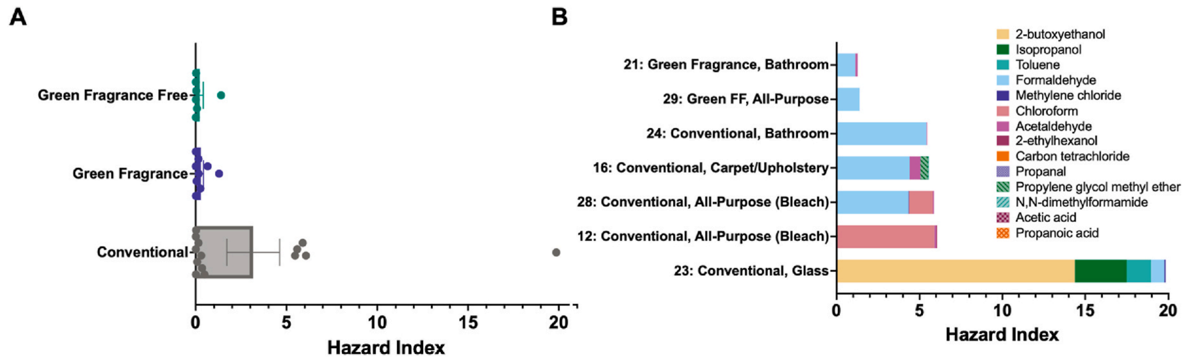

Fig. 5. Assessment of relative risk from products across groups and highest risk products. For each product, a hazard quotient was calculated for each VOC for which an exposure limit had been identified by dividing the quantified concentration by the exposure limit. A hazard index for each product was then calculated by adding together all hazard quotients for a given product. A) Average hazard index (± standard error) for conventional (n = 14), green (n = 8), green-fragrance free products (n = 7). B) Hazard index for products with hazard quotients above one and the contributing hazard quotients from the top five VOCs. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

highest calculated Hazard Index, 2-butoxyethanol, isopropanol, and toluene contributed to a large proportion of the hazard index, and in two bleach products tested, chloroform accounted for a large proportion of the risk (Fig. 5B). Due to the extremely low health-based exposure limit for formaldehyde, formaldehyde accounted for a high percentage of the associated product risk in several products, including those identified as green.

3.4. Comparison of VOCs emitted to ingredient disclosure and matching to regulatory or industry databases

Ingredient disclosure was assessed through labels on the package as well as information further disclosed on product manufacturers and retailers websites. Of the 23 products that were not identified as fragrance-free, 7 products listed fragrance as an ingredient on the product with no further disclosure, while 8 products disclosed some fragrance ingredients. About one fourth of emitted VOCs, 142 chemicals, were identified as chemicals used in fragrance, according to the International Fragrance Association, which includes both fragrance ingredients as well as non-fragrance ingredients like solvents, antioxidants, or preservatives, known as functional ingredients. Disclosed non-fragrance chemicals included ethanol, acetic acid, toluene and several glycol ethers (Table S3). Certain product categories, like glass cleaners, disclosed the use of solvent VOCs and often had high concentrations of these VOCs emitted from the products. Two products made on pack claims of being formaldehyde free and test results were below quantification limit or non-detect.

Out of the 530 detected VOCs just over half or 281 (53%) were listed in the ECHA C&L Inventory, 231 (44%) were listed on the public portion of the U.S. EPA TSCA inventory, and 91 (17%) were listed on the American Cleaning Institute cleaning product ingredient database.

4. Discussion

4.1. Air chamber methodology for the analysis of cleaning product emissions

Our study utilized air chambers to measure VOC emissions from two air fresheners and 28 cleaning products across several usage categories and product forms, to be representative of those likely used to clean within a household setting for a variety of cleaning tasks. The air chamber testing was designed to simulate product usage that would be relevant for potential VOC exposure for product usage, such that products were applied in their product forms the same way they would be applied during real world cleaning. Although the number of applications were lower than a real-world scenario given the small size of the chamber, the application rate was similar to rates estimated from a cleaning product use survey (Singer et al., 2006b). While this resulted in different application amounts, our results were generally similar when expressed on a μg/m^3 basis, μg/application unit and μg/g scenarios with the exception that air fresheners had far higher μg per gram emission factors. Naturally, the amount of product used is going to vary considerably between different users in real world scenarios. Also, while the form and category analysis for products tested here found no significant difference in air chamber concentrations (Figure S2), other studies indicate that spray product forms and cleaning with bleach products emits high levels of VOCs or concerning VOCs (Calderon et al., 2022; Loven et al., 2023). Our results for the conventional, green and green fragrance-free products were not statistically significant when considering emissions on a μg/g product scenario, although statistically significant differences were observed for VOC concentrations and μg emitted per application type. While on average, green and green fragrance-free products emitted lower numbers and concentrations of total VOCs in general and hazardous VOCs in particular, this finding is nuanced by the fact that some conventional products, including products that disclosed the presence of ingredients classified as hazardous, performed similarly in testing to some fragrance-free products.

While informative, the chamber testing does not fully replicate in home or workplace exposure scenarios where byproducts of cleaning product reactions can occur due to the presence of organic matter on surfaces and ozone in the air. In our study, carcinogenic chemicals chloroform and methylene chloride were detected in the air chamber after testing two bleach products, but whether these compounds were in the formulated product or generated in the chamber would require additional study. The chamber testing likely underestimates the real word exposure to disinfection byproducts given that bleach can form primary and secondary contaminants (Mattila et al., 2020). In a real word mopping scenario with a bleach product, more than ten chlorinated VOC species were identified (Arata et al., 2021). Other bleach cleaning scenarios detected high levels of carbon tetrachloride, chloroform and other halogenated VOCs (Odabasi, 2008). Additionally, a recent investigation of paired testing of VOC emissions in a home setting and in laboratory setting with an air chamber found general agreement between the two approaches, suggesting that chamber testing is a reasonable proxy for real world VOC exposure from cleaning products (Calderon et al., 2022).

Further, a data quality limitation applies to the identification of hazardous VOCs, creating an uncertainty in the assessment of potential health risk from cleaning product exposure. The identification of specific hazardous chemicals in the present study was based on both the DTSC Candidate Chemicals List and in the ECHA C&L Inventory. The DTSC Candidate Chemicals List is a compilation of toxicity assessments and serves as a metric for well-studied ingredients with identified hazards as well as a list of chemicals that companies are required in California to disclose, if intentionally added to products. The hazards in the ECHA C&L inventory contain hazard classifications that are both harmonized and non-harmonized, the latter meaning self-classified by the company (such as manufacturer or importer) notifying ECHA. The self-classified hazards are those identified by at least one company, whereby some identified hazards may not be applicable to all uses of the chemical from all suppliers. For the purposes of our analysis, these self-classifications were conservatively limited to a subset of endpoints that we considered relevant for cleaning product usage, specifically longer-term harm that could come from exposure to chemical product ingredients. While several of the hazardous VOCs emitted had evidence of respiratory toxicity, commonly seen in epidemiological studies of cleaning product use, many chemicals with other health harms such as neurotoxicity, carcinogenicity, and reproductive toxicity were also identified. Associations between cleaning product usage and potentially increased risk of non-respiratory adverse health outcomes needs further research.

The exposure limits analysis we conducted accounted for just under half of the total VOC emissions from products due to the hundreds of measured VOCs without published health benchmark values. The hazard index analysis across products was influenced by a few compounds, particularly formaldehyde, which has a very low health-based exposure limit. At the same time we consider it important to emphasize that occupational exposure limits for the same chemical across agencies can differ by orders of magnitude in part because many occupational exposure limits were set decades ago and may not be protective of worker health (Michaels and Barab, 2020).

4.2. Comparison of results with prior studies

These results are consistent with previous studies that found hundreds of different VOCs are emitted from products, the majority of which are not disclosed on ingredient labels (Steinemann, 2009). The overall VOC composition identified in different studies is ultimately a reflection of differences among the formulations of products in each study. Our findings differ from previous reports which found no significant differences between the number of VOCs and hazardous VOCs emitted from conventional and products classified as green (Nematollahi et al., 2018, 2019; Steinemann et al., 2021; Steinemann, 2009). However, previous analyses were limited only to the number of VOCs detected and not the concentration of VOCs emitted from the product since VOCs were assessed using headspace gas chromatography and not chamber testing. Further, these studies utilized different hazard list to identify chemicals of concern. We also observed an important similarity in that limonene was a prominent VOC in our study and in publications by Steinemann et al. Limonene was the sixth most commonly detected VOC in our analysis, detected in less than a third of products tested, and was among the top 10 VOC's contributing to total VOC emissions across all products, with about half from conventional, half from green fragrance and none from green fragrance-free products.

Emissions of VOCs from household products, especially terpenes such as limonene, can react with ozone present in the air at environmentally relevant concentrations and create secondary organic pollutants, including fine particulate matter and formaldehyde, both associated with adverse health effects (Coleman et al., 2008; Destaillats et al., 2006; Rosales et al., 2022; Singer et al., 2006a). Other ozone-reactive terpenes identified in our study were linalool, gamma terpineol, myrcene, linalyl acetate, alpha pinene and beta pinene. Recent investigations indicated that VOC emissions from consumer products equal contributions to total outdoor VOCs from vehicle emissions (McDonald et al., 2018). Additionally, VOCs from consumer products can themselves contribute to ozone formation creating secondary adverse health effects (Coggon et al., 2021).

Studies comparing real world usage of conventional versus green cleaning products found similar results as the present study of simulated product use. Reduced exposure to several hazardous VOCs including carcinogens, developmental and reproductive toxins, and endocrine disruptors, was observed in Latina women who switched from conventional to green cleaning products in an intervention study (Harley et al., 2021). In the same study, some women experienced exceedances in health-based exposure limits during conventional cleaning, but not during green cleaning. A recently published follow up to this study found that in chamber testing of conventional and green products, 75 percent of the highest VOC emissions came from conventional products (Calderon et al., 2022). In another study, the use of a bleach or quaternary ammonium compound based disinfectant and cleaner in a bathroom setting emitted more total VOCs than with the green disinfectant and cleaning product (Lindberg et al., 2021). Similarly, in the HOMEChem study which monitored whole house VOC emissions, emissions from mopping events with a bleach cleaner yielded more VOCs than cleaning with a product that the study defined as "all natural cleaning product with lemon-verbena scent", with nearly completely different emission profiles between the two (Arata et al., 2021). In a study of office floor cleaning in Europe, replacing conventional floor cleaners with a low-emissions floor cleaner reduced concentrations of limonene and formaldehyde (Norgaard et al., 2014).

Overall, findings from the present study highlight the wide variety of chemicals in cleaning product formulations and variations in VOC emissions. The large number of VOCs presents a challenge for identifying the VOCs that could potentially contribute to health harm both in direct users and in bystanders. More research is essential to identify the substances responsible for occupational and general population respiratory impacts (Clausen et al., 2020). Additionally, researching linking product interventions and exposure reductions to measurable improved health outcomes is limited and difficult to conduct.

Non-volatile compounds may also play an important role in health impacts from cleaning product use. Spray type products in particular have been implicated in causing respiratory harm, likely due to the production of droplets within the particle size range that can deposit in the respiratory system (Loven et al., 2019). Other specific chemicals implicated in increasing asthma are disinfectants including quaternary ammonium compounds, sodium hypochlorite, and gluteraldehyde, as well as fragrance chemicals eugenol and 3-carene listed by authoritative agencies as occupational asthmagens (Rosenman and Beckett, 2015). Recently, detection of quaternary ammonium compounds in breast milk was associated with use of disinfectants containing these compounds, with the highest exposure associated with use of spray products (Zheng et al., 2022).

4.3. Occupational exposure to cleaning product ingredients

Persons who clean professionally face increased exposure to VOCs emitted by cleaning products, including VOCs identified in the present study. Professional cleaning workers experience an elevated risk for the development or exacerbation of asthma and other respiratory harms and chronic health conditions (Oyer-Peterson et al., 2022). Chemical exposures in occupational settings could affect persons for whom cleaning professionally is a primary source of livelihood and employment (Michaels and Barab, 2020). Company-employed workers also are unlikely to be the persons making purchasing decisions on which products to use. Information on which cleaning products have safer chemical profiles both for VOC and non-VOC chemicals would be helpful both for cleaning professionals and individuals who clean in their own home. We emphasize that, of the VOCs detected in our study, most substances do not have published health-based reference limits, highlighting an essential research need to develop more comprehensive indoor air health standards.

4.4. Future research needs

Given the wide variety of chemicals emitted from different cleaning products, there is a strong need for additional study of respiratory chemical exposure and other health outcomes, in both children and adults. Studies documenting harm from cleaning product exposure often lack chemical specific resolution to identify the chemicals or mixtures of chemicals driving the adverse effect. In the absence of chemical-specific toxicity data, total VOC emissions and emissions of hazardous VOCs can be used as a metric for potential health harm. Future studies based on specific product use information or VOC monitoring may enable identification of the compounds most associated with harm.

Measuring VOC emissions from products in a test chamber is used as a proxy for exposure in cleaning scenarios. When comparing emissions contributing to exposure from green and conventional products in a real-world setting, reduced exposure to hazardous VOCs was observed when utilizing green cleaning products (Harley et al., 2021). Important distinctions were observed among different product type with respect to the chemical emissions. The use of bleach products was associated with elevated chloroform levels (Calderon et al., 2022), a result confirmed in our findings where chloroform was detected in VOC emissions from two bleach products. The one hypochlorous acid product had no detectable chloroform, and more testing of these disinfection products in real world conditions is warranted Testing an increased number of products would enable additional analysis of differences in emissions between product categories.

In published studies, sprays in particular have been implicated in causing the most adverse respiratory harm (Clausen et al., 2020). We noted that a single spray from different products could yield different amounts of cleaning substance emitted, likely based on the nozzle and manufacturing of the packaging. Assessing differences in the amount of product emitted between different yet similar products can have implications on end user exposure. On the other hand, the observed lower amount of VOC emissions in green products compared to conventional products in our study may be influenced by the fact that green products generally emitted less cleaning substance. We note that real-world exposure would be influenced by product efficacy (and perceived need to apply more or less of the cleaning substance), and the base condition of the area being cleaned as well as ventilation rates in the home, which can vary(Arata et al., 2021) Further research is needed on how best to conduct and translate chamber testing results to real-world scenarios.

4.5. Marketplace change

The increased consumer interest in product ingredient transparency and in certifications indicating products are safer for health or the environment has led to a new category of cleaning products. Our results indicate that green products tested in this study had reduced number and concentration of VOC and hazardous VOC emissions. A significant reduction in VOCs was observed for fragrance-free products indicating that these products offer potentially the greatest benefit in terms of reduced VOC exposure. U.S. EPA Safer Choice offers a specific certification label for fragrance-free products but to date no program requires or discloses the results from VOC emissions testing (U.S. EPA, 2016).

Increased ingredient disclosure prompted both by consumer advocacy and legislative efforts has provided consumers, researchers, and regulators more details on what ingredients are being added to products. VOC testing in the present study, and previous studies, detected hundreds of chemicals not disclosed on product labels or industry ingredient disclosure databases indicating a shortcoming in disclosure requirements for cleaning products. Our results also identified dozens of chemicals emitted from products that are part of a list of chemicals requiring disclosure under the state of California legislation if intentionally added to the product.

5. Conclusion

On average, even considering the large variability across the group of 30 products tested here, green cleaning products emitted fewer VOCs and fewer hazardous VOCs than conventional products, and overall fragrance-free products had significantly lower emissions. Our analysis also found a large variation between products with respect to the concentration of emitted VOCs, number of VOCs and the relative hazard index. The choice of green cleaning products, especially those that are fragrance-free, may offer consumers and occupational cleaners a way to reduce VOC emissions and reduce exposure to chemicals emitted from cleaning products.

Author contributions

SLG, AMT, SAS, NSL and DQA participated in experimental design and product selection. SLG, AMT, SAS and DQA analyzed the data. SLG, AMT, SAS, OVN and DQA wrote and edited the manuscript.

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Declaration of competing interest

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Data availability

Data tables of all chamber testing results provided in supplemental files.

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Appendix A. Supplementary data

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