Title
Large Emissions of Low Volatility Siloxanes during Residential Oven Use

Journal
Environmental Science and Technology

Authors
Katz, Erin F.
Lunderberg, David M.
Brown, Wyatt L.
et al.

Publication Date
2021-07-01

Peer reviewed

Large Emissions of Low-Volatility Siloxanes During Residential Oven Use

Erin F. Katz[1,2*], David M. Lunderberg[1,2], Wyatt L. Brown[3], Douglas A. Day[3], Jose L. Jimenez[3], William W Nazaroff[4], Allen H. Goldstein[2,4*], Peter F. DeCarlo[5*]

1. Department of Chemistry, University of California, Berkeley, California 94720, United States
2. Department of Environmental Science, Policy, and Management, University of California, Berkeley, California 94720, United States
3. Department of Chemistry and CIRES, University of Colorado, Boulder, Colorado 80309, United States
4. Department of Civil and Environmental Engineering, University of California, Berkeley, California 94720, United States
5. Department of Environmental Health and Engineering, Johns Hopkins University, Baltimore, Maryland 21218, United States

* Indicates corresponding authors

Abstract

Cooking is a source of airborne particles indoors and outdoors. A field study at a residential test house (HOMEChem) included two Thanksgiving-style cooking experiments involving prolonged use of an oven with light use history. Large enhancements of airborne low-volatility siloxanes were observed by three in-situ particle-phase instruments: a high-resolution aerosol mass spectrometer (HR-AMS), semivolatile thermal desorption aerosol gas chromatograph (SV-TAG), and extractive electrospray ionization mass spectrometer (EESI-MS). The combination of these instruments permits quantitatively analyzing time-dependent processes and fates over a wide volatility range with high chemical specificity. Cumulatively, 17 mg and 8.5 mg of bulk siloxane material was emitted indoors and observed in airborne particles during the first and second Thanksgiving experiments, respectively; a peak 5-minute average siloxane concentration of 58 $\mu g/m^3$ was measured. Cyclic siloxanes D10-D18 were quantified, and D17 and D16 were the most abundant. We infer that heating of silicone materials inside the oven caused volatilization of cyclic siloxanes and cooler temperatures away from the oven resulted in condensation. Low-volatility siloxanes comprised a surprisingly high fraction of the total emitted submicron particle mass: 18% and 9% during the first and second Thanksgiving experiments, respectively. We estimate ~75% of the low-volatility siloxane mass was ventilated outdoors.

Introduction

Indoor cooking causes emissions of suspended particulate matter (PM) and volatile organic compounds (VOC).[1] Cooking emissions have been analyzed in laboratory[2-3] as well as indoor[4-5] and outdoor field studies.[6-7] Particles emitted during cooking are mostly organic, and molecule markers of cooking emissions include oleic acid, levoglucosan, and cholesterol.[1] Emissions of semivolatile organic compounds (SVOCs) from indoor surface reservoirs and building materials have been identified as an additional source of particle mass during residential cooking.[8]

Methyl siloxanes ("siloxanes") have repeating $-Si(CH_3)_2-O-$ units and may be cyclic, linear, or branched. Cyclic volatile methyl siloxanes, such as D4, D5, and D6 (which have 4, 5, and 6 chain units, respectively), have gained attention as environmental contaminants due to their prevalence in consumer products.[9-10] In particular, D5 has been observed as the most abundant siloxane in urban atmospheres,[11-12] indoor environments,[13-14] and wastewater treatment plants[15] presumably due to personal care product use. Volatile siloxanes have the potential for long range

atmospheric transport[16] and bioaccumulation.[17] Evidence from laboratory studies suggests that their oxidation leads to secondary aerosols.[18] Migration of siloxanes from food contact materials (e.g., silicone baking molds) to food has been observed,[19-20] and an action limit of 60 mg of siloxanes per kg of food has been proposed in Norway for cyclic and linear siloxanes.[21] The European Union has recognized D4 and D5 as "very persistent and very bioaccumulative" under the REACH initiative and aims to reduce emissions of siloxanes in consumer and professional products.[22] Laboratory and ecological studies suggest that the health risks associated with exposure to volatile siloxanes are low.[23-24]

The HOMEChem study was designed to identify and quantify emissions associated with human activity in a real indoor environment through scripted experiments with intensive analytical monitoring.[25] Because the majority of peoples' time is spent indoors,[26] it is important to study the chemistry of indoor environments to assess VOC and PM exposure and health risks. Here, we present an investigation of a surprising finding from Thanksgiving holiday style experiments conducted at HOMEChem, which involved prolonged operation of a residential oven with a light history of prior use. High concentrations of large, low-volatility siloxanes with 10 chain units or more (D10 and larger) were observed by in-situ instruments.[8, 27] Prior HOMEChem studies have also reported low-volatility siloxanes, but focused on emissions from surface reservoirs and instrument/method development.[8, 27] Here, we present a detailed report of the emission and fate of low-volatility siloxanes that occurred during the two HOMEChem Thanksgiving experiments.

Materials and Methods

The HOMEChem (House Observations of Microbial and Environmental Chemistry) study (June 1-28, 2018) involved controlled and repeated experiments that simulated household activities.[25] The study was conducted with an internal air recirculation rate of 8 h^{-1}, which promoted rapid mixing, and exchange rate with outdoor air of 0.5 ± 0.1 h^{-1}.[25] The kitchen had a propane gas stove and oven (free-standing gas range JGBS04BEMWH, General Electric, Boston, MA, Figure S1), which had been installed ten years earlier and had been used once, two months prior to the campaign. Emission rates and concentrations of PM are high during the initial use of ovens (and toaster ovens) and decline after repeated use.[28-29] Wallace and Ott [29] showed that PM number concentrations declined by 50% after the first use of an oven. On June 5, the HOMEChem oven was operated at 260 ºC for one hour while all house doors and windows were open; no analytical monitoring occurred during this period. On June 8, the oven operated at 177 ºC for 60 minutes to bake a frozen lasagna. In this report, results from two HOMEChem Thanksgiving experiments are presented (June 18 and June 27). The same cooking methods were used each day. Thanksgiving experiments involved stove and oven use (204 °C to 232 °C) for ~ 6 hours. Turkey, stuffing, pie, gravy, cranberry sauce, brussels sprouts, and sweet potato casserole were prepared by 4 researchers. Cooking utilized disposable aluminum, glass, metal, and Teflon non-stick cookware; no silicone baking molds or utensils were used. Instrumentation utilized in this study include a scanning mobility particle sizing instrument (SMPS Model 3936: TSI 3081 long differential mobility analyzer and TSI 3775 condensation particle counter), a high-resolution time-of-flight aerosol mass spectrometer (HR-AMS),[30-31] a semivolatile thermal desorption aerosol gas chromatograph (SV-TAG),[32-33] and an extractive electrospray ionization time-of-flight mass spectrometer (EESI-MS).[27, 34]

Instrument details are given in the Supplemental Information (SI). Briefly, the HR-AMS measured bulk siloxane material in the particle phase. The fragmentation involved in detection prevents molecular specificity. The summed signal of siloxane ions ($C_wH_xO_ySi_z^+$) was converted

to a mass concentration using the relative ionization efficiency (*RIE*) applied to organics (1.4) and assuming a collection efficiency (*CE*) of 1.0 (uncertainty = 39%). Additional details are presented in Katz et al. [35]

The SV-TAG characterized gas plus particle and particle-only concentrations for D10-D18 (uncertainty = 15%). The SV-TAG sampling scheme involves 20 minutes of sample collection followed by analysis to provide hourly measurements. Siloxane species were quantified using the nearest alkane in retention time as a calibration surrogate.[8] Intercomparison between the HR-AMS bulk siloxane concentration and SV-TAG sum of D13-D18 (particle-only) indicates good agreement between the independently calibrated instruments (Figure S3). A slope of 1.15 (R^2 = 0.87) was calculated for a linear fit of HR-AMS versus SV-TAG data when excluding an outlying point where we infer siloxanes D19 and larger more substantially contributed to the signal. (The slope was 1.51 (R^2 = 0.88) when including that point.)

The EESI-MS uses soft (extractive electrospray) ionization time-of-flight mass spectrometry to detect organic species in airborne particles and provides near-molecular identification.[34] D13-D17 were detected by EESI-MS.[27] The EESI-MS raw signal was converted to a mass concentration by scaling to the SV-TAG data (Figure S4). The EESI-MS and HR-AMS measurements are presented as 5-minute averages. The SMPS was used to calculate total particle mass emissions (density = 1.0 g/cm^3) and number size distributions. Unit density was shown to be appropriate by Katz et al. [35] The SMPS detected particles with mobility diameters of 15-660 nm.

Siloxane emissions were calculated using Equation 1, where E is the total emitted mass (µg), ΔT is event duration (h), V is the interior house volume (235 m^3), L is the combined loss rate due to air exchange and deposition (0.66 h^{-1}, calculated by fitting the bulk siloxane decay curves at the end of each Thanksgiving day experiment when emission was assumed to be zero, (Figure 1)), C_{avg} is the average concentration throughout ΔT (µg/m^3), and $(C_2 - C_1)$ is the change in concentration from the start of the event (C_1) to the end of the event (C_2). Background (unoccupied) concentrations of siloxanes were 0.007 $\pm$ 0.002 µg/m^3 on average (HR-AMS) and at or below the detection limit (SV-TAG).

$$E = V \times (C_2 - C_1) + (V \times L \times \Delta T \times C_{avg}) \quad (1)$$

Results and Discussion

Surprisingly high concentrations of siloxanes in particulate matter were observed during HOMEChem Thanksgiving experiments (Figure 1). The HR-AMS, which measured bulk siloxane material in airborne particles, reported peak 5-minute average siloxane concentrations of 58 µg/m^3 and 25 µg/m^3 during the first and second Thanksgiving experiments, respectively. Considering the duration of both Thanksgiving experiments (20 hours of data, 9:05-19:05 on each day), the average airborne siloxane concentration was 8.0 µg/m^3. Siloxane D17 was the most abundant (2.2 µg/m^3 on average), followed by D16 (1.4 µg/m^3 on average) (Table S2). Siloxanes D13 through D18 were observed primarily in the particle phase, while D10 through D12 had considerable gas-phase fractions (Table 1). Total siloxane emissions were 17 mg and 8.5 mg, which were 18% and 9% of the total particle mass emitted during the first and second Thanksgiving experiments, respectively. The emissions of D17 and D18 were higher on the first Thanksgiving experiment (4200 and 1300 µg, respectively) compared to the second (2700 and 300 µg, respectively), whereas the other siloxane emissions were consistent between experiments (Table 1).

Prior studies reporting on siloxanes indicate their emission may be a feature of newer products. For example, Fromme et al. [20] measured D3-D9 in indoor air during baking with silicone

molds. Baking with newer molds resulted in higher airborne concentrations of D3-D9 (up to ~2000 μg/m^3); concentrations declined after repeated use.[20] Helling et al. [19] reported high levels of D6-D18 siloxanes in a 10-times used pizza mold, whereas a 1700-times used pizza mold had no detectable cyclic siloxanes. Schripp et al. [28] reported higher emission of ultrafine particles after the first use of household appliances such as toaster ovens. Similarly, the elevated siloxane levels observed during HOMEChem Thanksgiving experiments may be a feature of the lightly used oven. Annis and Cicero [36] report that the "burning off of insulation oils and binders" during the first few uses of an oven's self-cleaning cycle is expected. During another study that utilized SV-TAG (at a site designated as H2, a normally occupied residence),[4] D11-D18 were not observed at elevated levels during oven use (Figure S5). The absence of low volatility siloxanes at H2 could be because the oven was heavily used prior to that study, so complete removal of siloxanes might have been achieved. Another factor to consider is the long duration of oven use (~6 h) during the HOMEChem Thanksgiving experiments, which may have provided the opportunity for emission and condensation of low-volatility siloxanes. Additional controlled studies are needed to determine how appliance use patterns affect indoor siloxane emissions.

The fate of siloxanes emitted during Thanksgiving experiments was estimated by considering two ultimate outcomes: attachment/deposition to indoor surfaces or transfer to outdoors via ventilation. To estimate these fates, we consider rate constants for the following first-order processes: molecular deposition/attachment to surfaces or preexisting particles, deposition of particles to surfaces, and removal to outdoors via ventilation. The D17 diffusion coefficient was used for calculations, as this siloxane was the most abundant. Molecular D17 was estimated to be lost from air to indoor surfaces and airborne particles at rates of ~ 3 h^{-1} and 450 h^{-1}, respectively (SI Section 2). The loss rate to indoor surfaces was calculated using the same parameters as Wang et al. [37] and the loss rate to particles was calculated using the Fuchs-Sutugin expression for condensation flux, as presented in Seinfeld and Pandis [38] using the particle number size distribution measured by SMPS during the time of peak siloxane concentration. Assuming that losses are first-order processes operating in parallel, ~99% of the molecular D17 is expected to have condensed onto airborne particles and 1% would have been lost to indoor surfaces. The subsequent fate of particle-phase siloxanes is either deposition to indoor surfaces or ventilation to outdoors. The difference between the empirically derived total siloxane loss rate (L = 0.66 h^{-1}) and loss rate due to ventilation (k_v = 0.5 h^{-1}) provides an estimate of siloxane loss rate due to deposition on indoor surfaces, k_s = 0.16 h^{-1}. Xu et al. [39] have reported similar deposition rates for tobacco smoke particles indoors. Assuming that ventilation and particle deposition are first-order processes operating in parallel, 75% of the bulk siloxane mass in PM would have been ventilated to outdoors and 25% would have deposited onto indoor surfaces. Combining these results, we estimate ~75% of the low-volatility siloxanes emitted during Thanksgiving experiments would have escaped to the outdoor atmosphere with the remaining 25% depositing on indoor surfaces.

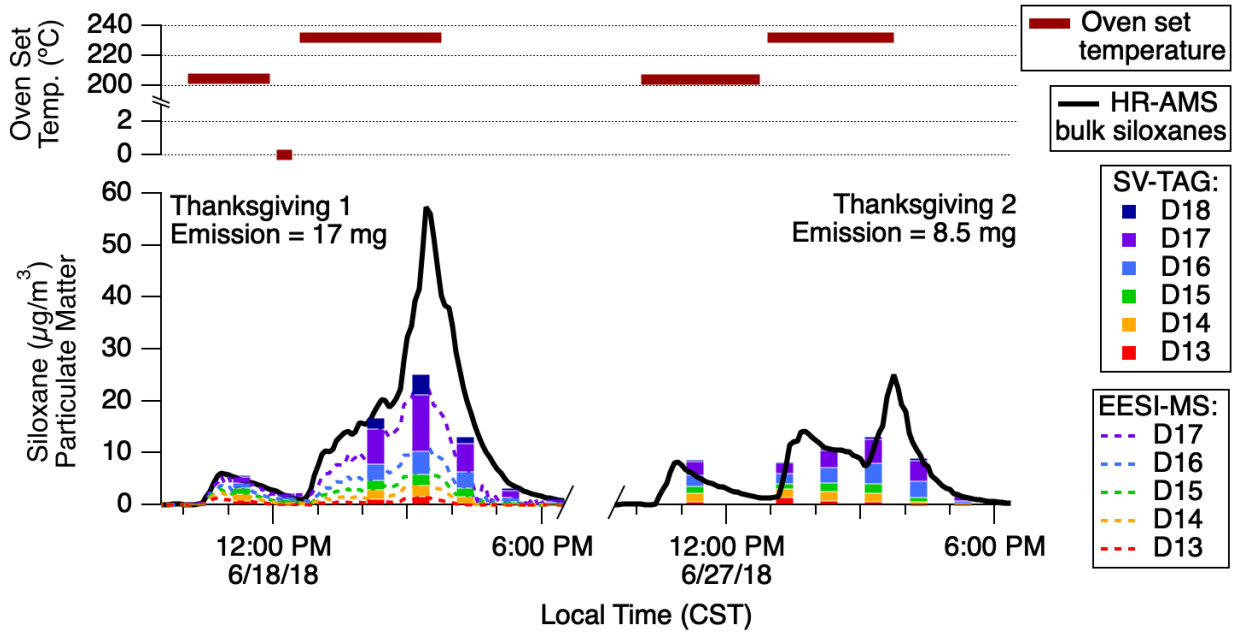

Figure 1*: Concentration of siloxanes in airborne particles measured by SV-TAG, EESI-MS, and HR-AMS and the oven set-point temperature during HOMEChem experiments Thanksgiving 1 (June 18, 2018) and 2 (June 27, 2018). The HR-AMS measured bulk siloxane material ($C_wH_xO_ySi_z^+$ ion fragments), whereas the SV-TAG and EESI-MS measured speciated cyclic methyl siloxanes (D13 to D18). All data shown are for particle-phase only. The SV-TAG and HR-AMS were independently calibrated (uncertainties = 15% and 39%, respectively), and the EESI-MS response was scaled to the SV-TAG measurements. The bulk siloxane mass emitted is shown for each Thanksgiving experiment.*

Table 1*: Fraction in the particle phase (F_P) for speciated siloxanes and cumulative mass emitted during each Thanksgiving experiment.* [a]

	F_P (Thanksgiving 1 only)	Mass Emitted (particle, µg)	
		Thanksgiving 1	Thanksgiving 2
D10	0.15 ± 0.09	20	31
D11	0.33 ± 0.16	26	34
D12	0.59 ± 0.14	166	118
D13	0.84 ± 0.12	500	500
D14	0.94 ± 0.10	1100	1100
D15	predominantly particle	1200	1000
D16	predominantly particle	2100	2300
D17	predominantly particle	4200	2700
D18	predominantly particle	1300	300
Bulk Siloxane	-	17,000	8500
Total Mass	-	96,000	94,000

[a] Fraction in the particle phase (F_P) values refer to population averages and standard deviations. All mass values are shown in µg. Speciated emissions are from SV-TAG measurements, and the

bulk siloxane emission is from HR-AMS measurements. Total submicron particle mass emission is also reported (SMPS measurement with a density of 1.0 g/cm^3 used).[35] Emissions were calculated over a 10-hour duration for each experiment (9:05-19:05 on each day).

The HR-AMS and SV-TAG agreed within uncertainty estimates. During the morning of June 18, bulk siloxanes (HR-AMS) and sum of D13-D18 (SV-TAG) particle concentration agreed well and reached a concentration of ~5 µg/m^3 (Figure 1). In the afternoon, when the oven set-point temperature increased from 204 °C to 232 °C, concentrations increased and the HR-AMS reported about 1.5 times the siloxane concentration as the SV-TAG sum of D13-D18 averaged over the same sampling period. This discrepancy is likely attributable to siloxanes in addition to D13-D18, such as larger siloxanes that are measurable by the HR-AMS and fragment to produce signal at lower *m/z* but are difficult to detect and quantify with other techniques. At least D19 and D20 were qualitatively observed by SV-TAG.[8] The SV-TAG chromatograms suggest small influence from linear siloxanes (~1-10% the signal of cyclic siloxanes). During the second Thanksgiving experiment (June 27), better agreement was observed between the HR-AMS and SV-TAG throughout the day, possibly because of lower emissions of larger cyclic siloxanes such as D19 and D20. The EESI-MS agreed temporally with the HR-AMS siloxane signal during the first Thanksgiving experiment, providing further confirmation of these observations. The EESI-MS siloxane analysis was not available for the second Thanksgiving experiment.

The origin of these low volatility siloxanes is not certain. Building materials such as caulk and sealants contain silicone polymers,[10, 40] and were likely present inside and around the oven. Heat resistant silicone polymers used as lubricants, sealants, and heat-transfer fluids for protecting electronics were likely present inside of the oven, which we believe to be the most likely source.[8, 41] Cyclic siloxanes in silicone materials, particularly silicone baking molds, are considered "non-intentionally added substances".[19] We infer that heating the oven interior to a high temperature for an extended period drove the volatilization of cyclic siloxanes from silicone materials, and, once outside of the warm oven interior or immediate vicinity, the cooler temperatures led to condensation. Another possible source is thermal decomposition of polydimethylsiloxane (PDMS, possibly present in the oven interior as a sealant), which has been shown to produce cyclic siloxanes.[42]

The concentration time series of siloxanes and particle-phase organics followed similar temporal trends, although the ratio of siloxanes to organics varied throughout the day depending on the cooking event (Figure 2a). Siloxanes and organics had overlapping particle size distributions during Thanksgiving experiments (Figure 2b), indicating they were internally mixed. The siloxane size distribution was slightly shifted towards smaller vacuum aerodynamic diameters (D_{va}) relative to organics.

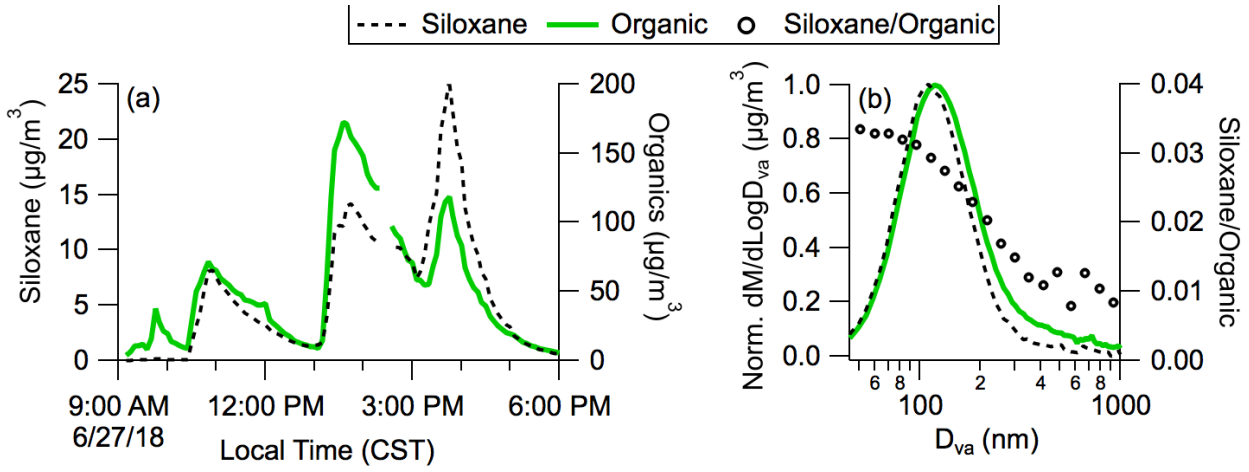

Figure 2*: (a) Organic and siloxane particle mass concentration versus time (measured by the HR-AMS) during the second Thanksgiving experiment. (b) Left axis: organic and siloxane normalized mass distribution versus vacuum aerodynamic diameter (D_{va}). Right axis: siloxane/organic ratio as a function of D_{va}. Organics are a proxy for total submicron particle concentration during HOMEChem cooking experiments. Not that the siloxane signal in (b) is the sum of select unit mass resolution ions (m/z>147) and does not represent the total siloxane fraction.*

During other HOMEChem cooking experiments of briefer duration that did not involve prolonged oven use, only slight enhancements (<1 µg/m^3) in siloxane concentration were observed (Figure S6). These low concentration observations could have originated from surface reservoirs of cooking organic aerosols,[8] heating cookware, or the toaster oven. During the oven use event on June 8 to bake a frozen lasagna, a low concentration siloxane enhancement was observed (5-min maximum of ~0.4 µg/m^3 and cumulative emissions of 70 µg over 1.5 hours).

HOMEChem and other indoor field studies provide insights to the complex processes that contribute to aerosol mass during cooking in real indoor environments.[4] This work, in addition to prior reports [8, 27] highlights the potential for non-food and non-fuel related chemicals to contribute aerosol mass during indoor cooking and motivates future studies to consider the health effects of inhaling these compounds in airborne particles.

Acknowledgements

We acknowledge Paula Olsiewski, Ph.D. and funding from the Alfred P. Sloan Foundation. We thank Atila Novoselac, Ph.D. and UT Austin for hosting the HOMEChem project, and Marina Vance, Ph.D. and Delphine Farmer, Ph.D. for leading the project.

This work was funded by grants from the Sloan Foundation Chemistry of Indoor Environments program (G-2019-12301, 2016-7173, 2019-12444). JLJ, DAD, and WLB thank a Cooperative Institute for Research in Environmental Sciences (CIRES) Innovative Research Program (IRP) grant, and a CIRES Fellowship for WLB.

Supplemental Information

The supplemental information contains images of the oven, additional instrument details, instrument calibration details and figures, chromatograms from the H2 field campaign oven use event, a figure from another HOMEChem cooking experiment with shorter duration oven use, SV-

TAG siloxane concentrations for each Thanksgiving experiment, and equations and parameters used to estimate the siloxane fate.

References

1. Abdullahi, K. L.; Delgado-Saborit, J. M.; Harrison, R. M., Emissions and Indoor Concentrations of Particulate Matter and its Specific Chemical Components from Cooking: A Review. *Atmos. Environ.* **2013,** *71*, 260—294.
2. Klein, F.; Farren, N. J.; Bozzetti, C.; Daellenbach, K. R.; Kilic, D.; Kumar, N. K.; Pieber, S. M.; Slowik, J. G.; Tuthill, R. N.; Hamilton, J. F.; Baltensperger, U.; Prévôt, A. S. H.; El Haddad, I., Indoor terpene emissions from cooking with herbs and pepper and their secondary organic aerosol production potential. *Sci. Rep.* **2016,** *6* (1), 36623.
3. Reyes-Villegas, E.; Bannan, T.; Le Breton, M.; Mehra, A.; Priestley, M.; Percival, C.; Coe, H.; Allan, J. D., Online Chemical Characterization of Food-Cooking Organic Aerosols: Implications for Source Apportionment. *Environ. Sci. Technol.* **2018,** *52* (9), 5308-5318.
4. Kristensen, K.; Lunderberg, D. M.; Liu, Y.; Misztal, P. K.; Tian, Y.; Arata, C.; Nazaroff, W. W.; Goldstein, A. H., Sources and dynamics of semivolatile organic compounds in a single-family residence in northern California. *Indoor Air* **2019,** *29* (4), 645-655.
5. Patel, S.; Sankhyan, S.; Boedicker, E. K.; DeCarlo, P. F.; Farmer, D. K.; Goldstein, A. H.; Katz, E. F.; Nazaroff, W. W.; Tian, Y.; Vanhanen, J.; Vance, M. E., Indoor Particulate Matter during HOMEChem: Concentrations, Size Distributions, and Exposures. *Environ. Sci. Technol.* **2020,** *54* (12), 7107-7116.
6. Mohr, C.; DeCarlo, P. F.; Heringa, M. F.; Chirico, R.; Slowik, J. G.; Richter, R.; Reche, C.; Alastuey, A.; Querol, X.; Seco, R.; Penuelas, J.; Jimenez, J. L.; Crippa, M.; Zimmermann, R.; Baltensperger, U.; Prevot, A. S. H., Identification and quantification of organic aerosol from cooking and other sources in Barcelona using aerosol mass spectrometer data. *Atmospheric Chemistry and Physics* **2012,** *12* (4), 1649-1665.
7. Allan, J. D.; Williams, P. I.; Morgan, W. T.; Martin, C. L.; Flynn, M. J.; Lee, J.; Nemitz, E.; Phillips, G. J.; Gallagher, M. W.; Coe, H., Contributions from transport, solid fuel burning and cooking to primary organic aerosols in two UK cities. *Atmos. Chem. Phys.* **2010,** *10* (2), 647-668.
8. Lunderberg, D. M.; Kristensen, K.; Tian, Y.; Arata, C.; Misztal, P. K.; Liu, Y.; Kreisberg, N.; Katz, E. F.; DeCarlo, P. F.; Patel, S.; Vance, M. E.; Nazaroff, W. W.; Goldstein, A. H., Surface Emissions Modulate Indoor SVOC Concentrations through Volatility-Dependent Partitioning. *Environ. Sci. Technol.* **2020,** *54* (11), 6751-6760.
9. Capela, D.; Alves, A.; Homem, V.; Santos, L., From the shop to the drain - Volatile methylsiloxanes in cosmetics and personal care products. *Environment International* **2016,** *92-93*, 50-62.
10. Horii, Y.; Kannan, K., Survey of Organosilicone Compounds, Including Cyclic and Linear Siloxanes, in Personal-Care and Household Products. *Archives of Environmental Contamination and Toxicology* **2008,** *55* (4), 701.
11. Coggon, M. M.; McDonald, B. C.; Vlasenko, A.; Veres, P. R.; Bernard, F.; Koss, A. R.; Yuan, B.; Gilman, J. B.; Peischl, J.; Aikin, K. C.; DuRant, J.; Warneke, C.; Li, S.-M.; de Gouw, J. A., Diurnal Variability and Emission Pattern of Decamethylcyclopentasiloxane (D5) from the Application of Personal Care Products in Two North American Cities. *Environ. Sci. Technol.* **2018,** *52* (10), 5610-5618.

12. Gallego, E.; Perales, J. F.; Roca, F. J.; Guardino, X.; Gadea, E., Volatile methyl siloxanes (VMS) concentrations in outdoor air of several Catalan urban areas. *Atmos. Environ.* **2017,** *155*, 108-118.
13. Tang, X.; Misztal, P. K.; Nazaroff, W. W.; Goldstein, A. H., Siloxanes Are the Most Abundant Volatile Organic Compound Emitted from Engineering Students in a Classroom. *Environ. Sci. Technol. Lett.* **2015,** *2* (11), 303-307.
14. Pieri, F.; Katsoyiannis, A.; Martellini, T.; Hughes, D.; Jones, K. C.; Cincinelli, A., Occurrence of linear and cyclic volatile methyl siloxanes in indoor air samples (UK and Italy) and their isotopic characterization. *Environment International* **2013,** *59*, 363-371.
15. Capela, D.; Ratola, N.; Alves, A.; Homem, V., Volatile methylsiloxanes through wastewater treatment plants - A review of levels and implications. *Environment International* **2017,** *102*, 9-29.
16. McLachlan, M. S.; Kierkegaard, A.; Hansen, K. M.; van Egmond, R.; Christensen, J. H.; Skjøth, C. A., Concentrations and Fate of Decamethylcyclopentasiloxane (D5) in the Atmosphere. *Environ. Sci. Technol.* **2010,** *44* (14), 5365-5370.
17. Gobas, F. A. P. C.; Powell, D. E.; Woodburn, K. B.; Springer, T.; Huggett, D. B., Bioaccumulation of decamethylpentacyclosiloxane (D5): A review. *Environ. Toxicol. Chem.* **2015,** *34* (12), 2703-2714.
18. Wu, Y.; Johnston, M. V., Aerosol Formation from OH Oxidation of the Volatile Cyclic Methyl Siloxane (cVMS) Decamethylcyclopentasiloxane. *Environ. Sci. Technol.* **2017,** *51* (8), 4445-4451.
19. Helling, R.; Seifried, P.; Fritzsche, D.; Simat, T. J., Characterisation and migration properties of silicone materials during typical long-term commercial and household use applications: a combined case study. *Food Additives & Contaminants: Part A* **2012,** *29* (9), 1489-1500.
20. Fromme, H.; Witte, M.; Fembacher, L.; Gruber, L.; Hagl, T.; Smolic, S.; Fiedler, D.; Sysoltseva, M.; Schober, W., Siloxane in baking moulds, emission to indoor air and migration to food during baking with an electric oven. *Environment International* **2019,** *126*, 145-152.
21. Cederberg, T. L.; Krüger, L., Siloxanes in silicone products intended for food contact: Selected samples from the Norwegian market in 2016. National Food Institute. Technical University of Denmark. 2017.
22. ECHA Registry of SVHC intentions until outcome. https://echa.europa.eu/registry-of-svhc-intentions/-/dislist/name/-/sbm_expected_submissionFrom/-/sbm_expected_submissionTo/-/ecNumber/-/dte_withdrawnFrom/-/dte_withdrawnTo/-/casNumber/-/lec_submitter/-/prc_public_status/-/haz_detailed_concern/-/dte_intentionFrom/-/dte_intentionTo/-/dte_opinionFrom/-/dte_opinionTo/-/dte_adoptionFrom/17-04-1963/dte_adoptionTo/31-12-2038/dte_inclusionFrom/-/dte_inclusionTo/-/ (accessed February 8, 2021).
23. Decamethylcyclopentasiloxane (D5). *Toxicol. Ind. Health.* **2017,** *33* (1), 16-27.
24. Fairbrother, A.; Burton, G. A.; Klaine, S. J.; Powell, D. E.; Staples, C. A.; Mihaich, E. M.; Woodburn, K. B.; Gobas, F. A. P. C., Characterization of ecological risks from environmental releases of decamethylcyclopentasiloxane (D5). *Environ. Toxicol. Chem.* **2015,** *34* (12), 2715-2722.
25. Farmer, D. K.; Vance, M. E.; Abbatt, J. P. D.; Abeleira, A.; Alves, M. R.; Arata, C.; Boedicker, E.; Bourne, S.; Cardoso-Sandaña, F.; Corsi, R.; DeCarlo, P. F.; Goldstein, A. H.; Grassian, V. H.; Ruiz, L. H.; Jimenez, J. L.; Kahan, T. F.; Katz, E. F.; Mattila, J. M.; Nazaroff, W. W.; Novoselac, A.; O'Brien, R. E.; Or, V. W.; Patel, S.; Sankhyan, S.; Stevens, P. S.; Tian,

Y.; Wade, M.; Wang, C.; Zhou, S.; Zhou, Y., Overview of HOMEChem: House Observations of Microbial and Environmental Chemistry. *Environ. Sci.: Process. Impacts* **2019,** *21*, 1280-1300.
26. USEPA, Report to Congress on Indoor Air Quality: Volume 2. EPA/400/1-89/001C. US Environmental Protection Agency. Washington, DC, 1989.
27. Brown, W. L.; Day, D. A.; Stark, H.; Pagonis, D.; Krechmer, J. E.; Liu, X.; Price, D. J.; Katz, E. F.; DeCarlo, P. F.; Masoud, C. G.; Wang, D. S.; Ruiz, L. H.; Arata, C.; Lunderberg, D. M.; Goldstein, A. H.; Farmer, D. K.; Vance, M. E.; Jimenez, J. L., Real-time organic aerosol chemical speciation in the indoor environment using extractive electrospray ionization mass spectrometry. *Indoor Air* **2020,** *31* (1), 141-155.
28. Schripp, T.; Kirsch, I.; Salthammer, T., Characterization of particle emission from household electrical appliances. *Sci. Total Environ.* **2011,** *409*, 2534-2540.
29. Wallace, L.; Ott, W., Personal exposure to ultrafine particles. *J. Expo. Sci. Environ. Epidemiol.* **2011,** *21* (1), 20-30.
30. DeCarlo, P. F.; Kimmel, J. R.; Trimborn, A.; Northway, M. J.; Jayne, J. T.; Aiken, A. C.; Gonin, M.; Fuhrer, K.; Horvath, T.; Docherty, K. S.; Worsnop, D. R.; Jimenez, J. L., Field-Deployable, High-Resolution, Time-of-Flight Aerosol Mass Spectrometer. *Anal. Chem.* **2006,** *78* (24), 8281-8289.
31. Canagaratna, M. R.; Jayne, J. T.; Jimenez, J. L.; Allan, J. D.; Alfarra, M. R.; Zhang, Q.; Onasch, T. B.; Drewnick, F.; Coe, H.; Middlebrook, A.; Delia, A.; Williams, L. R.; Trimborn, A. M.; Northway, M. J.; DeCarlo, P. F.; Kolb, C. E.; Davidovits, P.; Worsnop, D. R., Chemical and microphysical characterization of ambient aerosols with the aerodyne aerosol mass spectrometer. *Mass Spectrom Rev* **2007,** *26* (2), 185-222.
32. Zhao, Y.; Kreisberg, N. M.; Worton, D. R.; Teng, A. P.; Hering, S. V.; Goldstein, A. H., Development of an In Situ Thermal Desorption Gas Chromatography Instrument for Quantifying Atmospheric Semi-Volatile Organic Compounds. *Aerosol Sci. Technol.* **2013,** *47* (3), 258-266.
33. Isaacman, G.; Kreisberg, N. M.; Yee, L. D.; Worton, D. R.; Chan, A. W. H.; Moss, J. A.; Hering, S. V.; Goldstein, A. H., Online derivatization for hourly measurements of gas- and particle-phase semi-volatile oxygenated organic compounds by thermal desorption aerosol gas chromatography (SV-TAG). *Atmos. Meas. Tech.* **2014,** *7* (12), 4417-4429.
34. Lopez-Hilfiker, F. D.; Pospisilova, V.; Huang, W.; Kalberer, M.; Mohr, C.; Stefenelli, G.; Thornton, J. A.; Baltensperger, U.; Prevot, A. S. H.; Slowik, J. G., An extractive electrospray ionization time-of-flight mass spectrometer (EESI-TOF) for online measurement of atmospheric aerosol particles. *Atmos. Meas. Tech.* **2019,** *12* (9), 4867-4886.
35. Katz, E. F.; Guo, H.; Campuzano-Jost, P.; Day, D. A.; Brown, W. L.; Boedicker, E.; Pothier, M.; Lunderberg, D. M.; Patel, S.; Patel, K.; Hayes, P. L.; Avery, A.; Ruiz, L. H.; Goldstein, A. H.; Vance, M. E.; Farmer, D. K.; Jimenez, J. L.; DeCarlo, P. F., Quantification of cooking organic aerosol in the indoor environment using Aerodyne aerosol mass spectrometers. *Aerosol Sci. Technol.* **2021**.
36. Annis, P. J.; Cicero, J. H. L., The Self-Cleaning Oven: A Source of Inside Air Contamination. *Home Economics Research Journal* **1981,** *9* (3), 232-239.
37. Wang, C.; Collins, D. B.; Arata, C.; Goldstein, A. H.; Mattila, J. M.; Farmer, D. K.; Ampollini, L.; DeCarlo, P. F.; Novoselac, A.; Vance, M. E.; Nazaroff, W. W.; Abbatt, J. P. D., Surface reservoirs dominate dynamic gas-surface partitioning of many indoor air constituents. *Sci. Adv.* **2020,** *6* (8), eaay8973.
38. Seinfeld, J. H.; Pandis, S. N., *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*. 3 ed.; John Wiley & Sons, Inc: Hoboken, New Jersey, 2016.

39. Xu, M.; Nematollahi, M.; Sextro, R. G.; Gadgil, A. J.; Nazaroff, W. W., Deposition of Tobacco Smoke Particles in a Low Ventilation Room. *Aerosol Sci. Technol.* **1994,** *20* (2), 194-206.
40. Dow DOWSIL(TM) 3-6096 Adhesive Technical Data Sheet. https://www.dow.com/en-us/document-viewer.html?ramdomVar=7771385646442966 48&docPath=/content/dam/dcc/documents/en-us/productdatasheet/80/80-39/80-3925-dowsil-3-6096-adhesive.pdf (accessed June 24, 2020).
41. Chichester, C., Development of High-Service Temperature Fluids. DuPont, Ed. dupont.co.uk, 2019 (accessed June 24, 2020).
42. Ballistreri, A.; Garozzo, D.; Montaudo, G., Mass spectral characterization and thermal decomposition mechanism of poly(dimethylsiloxane). *Macromolecules* **1984,** *17* (7), 1312-1315.

For Table of Contents Only: