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1 Large Emissions of Low-Volatility Siloxanes During Residential Oven Use

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17 Abstract

18 Cooking is a source of airborne particles indoors and outdoors. A field study at a residential
19 test house (HOMEChem) included two Thanksgiving-style cooking experiments involving
20 prolonged use of an oven with light use history. Large enhancements of airborne low-volatility
21 siloxanes were observed by three in-situ particle-phase instruments: a high-resolution aerosol mass
22 spectrometer (HR-AMS), semivolatile thermal desorption aerosol gas chromatograph (SV-TAG),
23 and extractive electrospray ionization mass spectrometer (EESI-MS). The combination of these
24 instruments permits quantitatively analyzing time-dependent processes and fates over a wide
25 volatility range with high chemical specificity. Cumulatively, 17 mg and 8.5 mg of bulk siloxane
26 material was emitted indoors and observed in airborne particles during the first and second
27 Thanksgiving experiments, respectively; a peak 5-minute average siloxane concentration of 58
28 $\mu\text{g}/\text{m}^3$ was measured. Cyclic siloxanes D10-D18 were quantified, and D17 and D16 were the most
29 abundant. We infer that heating of silicone materials inside the oven caused volatilization of cyclic
30 siloxanes and cooler temperatures away from the oven resulted in condensation. Low-volatility
31 siloxanes comprised a surprisingly high fraction of the total emitted submicron particle mass: 18%
32 and 9% during the first and second Thanksgiving experiments, respectively. We estimate ~75% of
33 the low-volatility siloxane mass was ventilated outdoors.
34

35 Introduction

36 Indoor cooking causes emissions of suspended particulate matter (PM) and volatile organic
37 compounds (VOC).¹ Cooking emissions have been analyzed in laboratory²⁻³ as well as indoor⁴⁻⁵
38 and outdoor field studies.⁶⁻⁷ Particles emitted during cooking are mostly organic, and molecule
39 markers of cooking emissions include oleic acid, levoglucosan, and cholesterol.¹ Emissions of
40 semivolatile organic compounds (SVOCs) from indoor surface reservoirs and building materials
41 have been identified as an additional source of particle mass during residential cooking.⁸

42 Methyl siloxanes (“siloxanes”) have repeating $-\text{Si}(\text{CH}_3)_2\text{-O}-$ units and may be cyclic,
43 linear, or branched. Cyclic volatile methyl siloxanes, such as D4, D5, and D6 (which have 4, 5,
44 and 6 chain units, respectively), have gained attention as environmental contaminants due to their
45 prevalence in consumer products.⁹⁻¹⁰ In particular, D5 has been observed as the most abundant
46 siloxane in urban atmospheres,¹¹⁻¹² indoor environments,¹³⁻¹⁴ and wastewater treatment plants¹⁵
47 presumably due to personal care product use. Volatile siloxanes have the potential for long range

48 atmospheric transport¹⁶ and bioaccumulation.¹⁷ Evidence from laboratory studies suggests that
49 their oxidation leads to secondary aerosols.¹⁸ Migration of siloxanes from food contact materials
50 (e.g., silicone baking molds) to food has been observed,¹⁹⁻²⁰ and an action limit of 60 mg of
51 siloxanes per kg of food has been proposed in Norway for cyclic and linear siloxanes.²¹ The
52 European Union has recognized D4 and D5 as “very persistent and very bioaccumulative” under
53 the REACH initiative and aims to reduce emissions of siloxanes in consumer and professional
54 products.²² Laboratory and ecological studies suggest that the health risks associated with exposure
55 to volatile siloxanes are low.²³⁻²⁴

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

67

68 **Materials and Methods**

69 The HOMEChem (House Observations of Microbial and Environmental Chemistry) study
70 (June 1-28, 2018) involved controlled and repeated experiments that simulated household
71 activities.²⁵ The study was conducted with an internal air recirculation rate of 8 h⁻¹, which
72 promoted rapid mixing, and exchange rate with outdoor air of 0.5 ± 0.1 h⁻¹.²⁵ The kitchen had a
73 propane gas stove and oven (free-standing gas range JGBS04BEMWH, General Electric, Boston,
74 MA, Figure S1), which had been installed ten years earlier and had been used once, two months
75 prior to the campaign. Emission rates and concentrations of PM are high during the initial use of
76 ovens (and toaster ovens) and decline after repeated use.²⁸⁻²⁹ Wallace and Ott²⁹ showed that PM
77 number concentrations declined by 50% after the first use of an oven. On June 5, the HOMEChem
78 oven was operated at 260 °C for one hour while all house doors and windows were open; no
79 analytical monitoring occurred during this period. On June 8, the oven operated at 177 °C for 60
80 minutes to bake a frozen lasagna. In this report, results from two HOMEChem Thanksgiving
81 experiments are presented (June 18 and June 27). The same cooking methods were used each day.
82 Thanksgiving experiments involved stove and oven use (204 °C to 232 °C) for ~ 6 hours. Turkey,
83 stuffing, pie, gravy, cranberry sauce, brussels sprouts, and sweet potato casserole were prepared
84 by 4 researchers. Cooking utilized disposable aluminum, glass, metal, and Teflon non-stick
85 cookware; no silicone baking molds or utensils were used. Instrumentation utilized in this study
86 include a scanning mobility particle sizing instrument (SMPS Model 3936: TSI 3081 long
87 differential mobility analyzer and TSI 3775 condensation particle counter), a high-resolution time-
88 of-flight aerosol mass spectrometer (HR-AMS),³⁰⁻³¹ a semivolatile thermal desorption aerosol gas
89 chromatograph (SV-TAG),³²⁻³³ and an extractive electrospray ionization time-of-flight mass
90 spectrometer (EESI-MS).^{27, 34}

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

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

97 The SV-TAG characterized gas plus particle and particle-only concentrations for D10-D18
98 (uncertainty = 15%). The SV-TAG sampling scheme involves 20 minutes of sample collection
99 followed by analysis to provide hourly measurements. Siloxane species were quantified using the
100 nearest alkane in retention time as a calibration surrogate.⁸ Intercomparison between the HR-AMS
101 bulk siloxane concentration and SV-TAG sum of D13-D18 (particle-only) indicates good
102 agreement between the independently calibrated instruments (Figure S3). A slope of 1.15 ($R^2 =$
103 0.87) was calculated for a linear fit of HR-AMS versus SV-TAG data when excluding an outlying
104 point where we infer siloxanes D19 and larger more substantially contributed to the signal. (The
105 slope was 1.51 ($R^2 = 0.88$) when including that point.)

106 The EESI-MS uses soft (extractive electrospray) ionization time-of-flight mass
107 spectrometry to detect organic species in airborne particles and provides near-molecular
108 identification.³⁴ D13-D17 were detected by EESI-MS.²⁷ The EESI-MS raw signal was converted
109 to a mass concentration by scaling to the SV-TAG data (Figure S4). The EESI-MS and HR-AMS
110 measurements are presented as 5-minute averages. The SMPS was used to calculate total particle
111 mass emissions (density = 1.0 g/cm³) and number size distributions. Unit density was shown to be
112 appropriate by Katz et al.³⁵ The SMPS detected particles with mobility diameters of 15-660 nm.

113 Siloxane emissions were calculated using Equation 1, where E is the total emitted mass
114 (μg), ΔT is event duration (h), V is the interior house volume (235 m³), L is the combined loss rate
115 due to air exchange and deposition (0.66 h⁻¹, calculated by fitting the bulk siloxane decay curves
116 at the end of each Thanksgiving day experiment when emission was assumed to be zero, (Figure
117 1)), C_{avg} is the average concentration throughout ΔT ($\mu\text{g}/\text{m}^3$), and $(C_2 - C_1)$ is the change in
118 concentration from the start of the event (C_1) to the end of the event (C_2). Background (unoccupied)
119 concentrations of siloxanes were $0.007 \pm 0.002 \mu\text{g}/\text{m}^3$ on average (HR-AMS) and at or below the
120 detection limit (SV-TAG).

$$121 \quad E = V \times (C_2 - C_1) + (V \times L \times \Delta T \times C_{avg}) \quad (1)$$

122 123 124 **Results and Discussion**

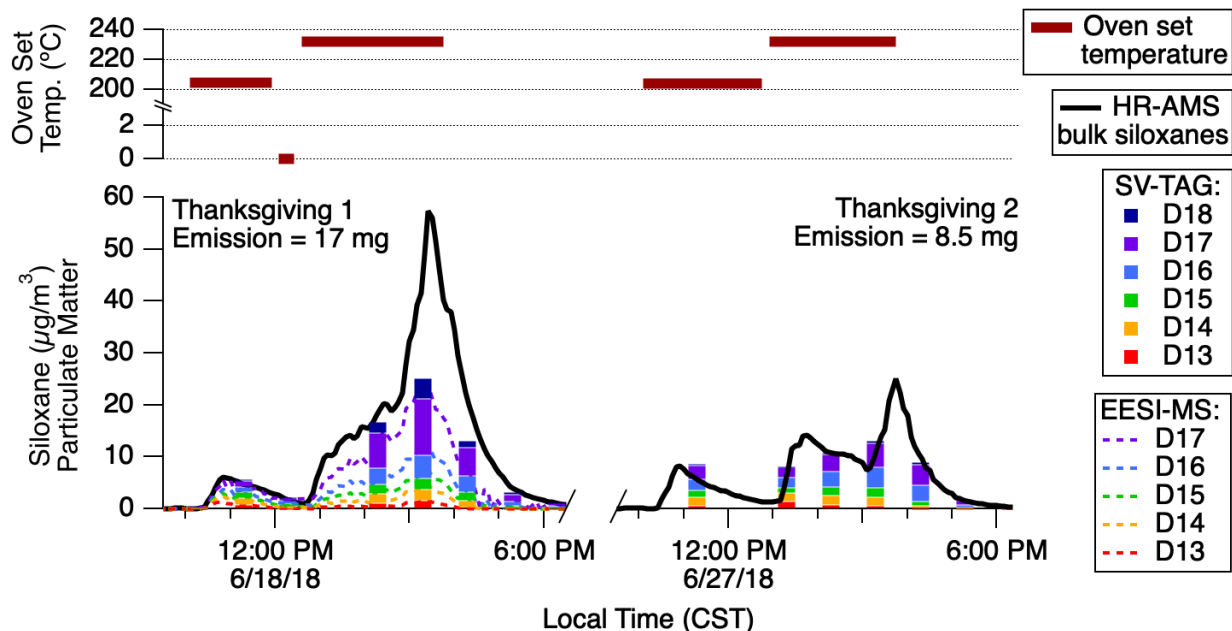
125 Surprisingly high concentrations of siloxanes in particulate matter were observed during
126 HOMEChem Thanksgiving experiments (Figure 1). The HR-AMS, which measured bulk siloxane
127 material in airborne particles, reported peak 5-minute average siloxane concentrations of 58 $\mu\text{g}/\text{m}^3$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

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

140 molds. Baking with newer molds resulted in higher airborne concentrations of D3-D9 (up to ~2000
141 $\mu\text{g}/\text{m}^3$); concentrations declined after repeated use.²⁰ Helling et al.¹⁹ reported high levels of D6-
142 D18 siloxanes in a 10-times used pizza mold, whereas a 1700-times used pizza mold had no
143 detectable cyclic siloxanes. Schripp et al.²⁸ reported higher emission of ultrafine particles after the
144 first use of household appliances such as toaster ovens. Similarly, the elevated siloxane levels
145 observed during HOMEChem Thanksgiving experiments may be a feature of the lightly used oven.
146 Annis and Cicero³⁶ report that the “burning off of insulation oils and binders” during the first few
147 uses of an oven’s self-cleaning cycle is expected. During another study that utilized SV-TAG (at
148 a site designated as H2, a normally occupied residence),⁴ D11-D18 were not observed at elevated
149 levels during oven use (Figure S5). The absence of low volatility siloxanes at H2 could be because
150 the oven was heavily used prior to that study, so complete removal of siloxanes might have been
151 achieved. Another factor to consider is the long duration of oven use (~6 h) during the
152 HOMEChem Thanksgiving experiments, which may have provided the opportunity for emission
153 and condensation of low-volatility siloxanes. Additional controlled studies are needed to determine
154 how appliance use patterns affect indoor siloxane emissions.

155 The fate of siloxanes emitted during Thanksgiving experiments was estimated by
156 considering two ultimate outcomes: attachment/deposition to indoor surfaces or transfer to
157 outdoors via ventilation. To estimate these fates, we consider rate constants for the following first-
158 order processes: molecular deposition/attachment to surfaces or preexisting particles, deposition
159 of particles to surfaces, and removal to outdoors via ventilation. The D17 diffusion coefficient was
160 used for calculations, as this siloxane was the most abundant. Molecular D17 was estimated to be
161 lost from air to indoor surfaces and airborne particles at rates of $\sim 3 \text{ h}^{-1}$ and 450 h^{-1} , respectively
162 (SI Section 2). The loss rate to indoor surfaces was calculated using the same parameters as Wang
163 et al.³⁷ and the loss rate to particles was calculated using the Fuchs-Sutugin expression for
164 condensation flux, as presented in Seinfeld and Pandis³⁸ using the particle number size distribution
165 measured by SMPS during the time of peak siloxane concentration. Assuming that losses are first-
166 order processes operating in parallel, ~99% of the molecular D17 is expected to have condensed
167 onto airborne particles and 1% would have been lost to indoor surfaces. The subsequent fate of
168 particle-phase siloxanes is either deposition to indoor surfaces or ventilation to outdoors. The
169 difference between the empirically derived total siloxane loss rate ($L = 0.66 \text{ h}^{-1}$) and loss rate due
170 to ventilation ($k_v = 0.5 \text{ h}^{-1}$) provides an estimate of siloxane loss rate due to deposition on indoor
171 surfaces, $k_s = 0.16 \text{ h}^{-1}$. Xu et al.³⁹ have reported similar deposition rates for tobacco smoke
172 particles indoors. Assuming that ventilation and particle deposition are first-order processes
173 operating in parallel, 75% of the bulk siloxane mass in PM would have been ventilated to outdoors
174 and 25% would have deposited onto indoor surfaces. Combining these results, we estimate ~75%
175 of the low-volatility siloxanes emitted during Thanksgiving experiments would have escaped to
176 the outdoor atmosphere with the remaining 25% depositing on indoor surfaces.

177



178
 179 **Figure 1:** Concentration of siloxanes in airborne particles measured by SV-TAG, EESI-MS, and
 180 HR-AMS and the oven set-point temperature during HOMEChem experiments Thanksgiving 1
 181 (June 18, 2018) and 2 (June 27, 2018). The HR-AMS measured bulk siloxane material ($C_wH_xO_ySi_z^+$
 182 ion fragments), whereas the SV-TAG and EESI-MS measured speciated cyclic methyl siloxanes
 183 (D13 to D18). All data shown are for particle-phase only. The SV-TAG and HR-AMS were
 184 independently calibrated (uncertainties = 15% and 39%, respectively), and the EESI-MS response
 185 was scaled to the SV-TAG measurements. The bulk siloxane mass emitted is shown for each
 186 Thanksgiving experiment.

187
 188 **Table 1:** Fraction in the particle phase (F_P) for speciated siloxanes and cumulative mass emitted
 189 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

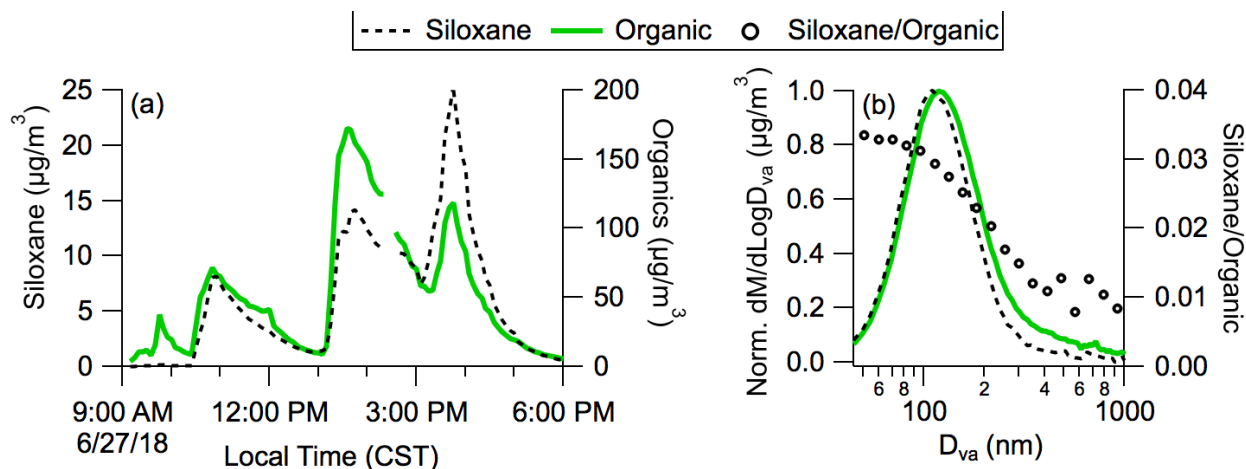
190 ^a Fraction in the particle phase (F_P) values refer to population averages and standard deviations.
 191 All mass values are shown in μg . Speciated emissions are from SV-TAG measurements, and the

192 bulk siloxane emission is from HR-AMS measurements. Total submicron particle mass emission
193 is also reported (SMPS measurement with a density of 1.0 g/cm³ used).³⁵ Emissions were
194 calculated over a 10-hour duration for each experiment (9:05-19:05 on each day).
195

196 The HR-AMS and SV-TAG agreed within uncertainty estimates. During the morning of
197 June 18, bulk siloxanes (HR-AMS) and sum of D13-D18 (SV-TAG) particle concentration agreed
198 well and reached a concentration of ~5 μg/m³ (Figure 1). In the afternoon, when the oven set-point
199 temperature increased from 204 °C to 232 °C, concentrations increased and the HR-AMS reported
200 about 1.5 times the siloxane concentration as the SV-TAG sum of D13-D18 averaged over the
201 same sampling period. This discrepancy is likely attributable to siloxanes in addition to D13-D18,
202 such as larger siloxanes that are measurable by the HR-AMS and fragment to produce signal at
203 lower *m/z* but are difficult to detect and quantify with other techniques. At least D19 and D20 were
204 qualitatively observed by SV-TAG.⁸ The SV-TAG chromatograms suggest small influence from
205 linear siloxanes (~1-10% the signal of cyclic siloxanes). During the second Thanksgiving
206 experiment (June 27), better agreement was observed between the HR-AMS and SV-TAG
207 throughout the day, possibly because of lower emissions of larger cyclic siloxanes such as D19
208 and D20. The EESI-MS agreed temporally with the HR-AMS siloxane signal during the first
209 Thanksgiving experiment, providing further confirmation of these observations. The EESI-MS
210 siloxane analysis was not available for the second Thanksgiving experiment.

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

222 The concentration time series of siloxanes and particle-phase organics followed similar
223 temporal trends, although the ratio of siloxanes to organics varied throughout the day depending
224 on the cooking event (Figure 2a). Siloxanes and organics had overlapping particle size
225 distributions during Thanksgiving experiments (Figure 2b), indicating they were internally mixed.
226 The siloxane size distribution was slightly shifted towards smaller vacuum aerodynamic diameters
227 (*D_{va}*) relative to organics.
228



229
 230 **Figure 2:** (a) Organic and siloxane particle mass concentration versus time (measured by the HR-
 231 AMS) during the second Thanksgiving experiment. (b) Left axis: organic and siloxane normalized
 232 mass distribution versus vacuum aerodynamic diameter (D_{va}). Right axis: siloxane/organic ratio
 233 as a function of D_{va} . Organics are a proxy for total submicron particle concentration during
 234 HOMEChem cooking experiments. Not that the siloxane signal in (b) is the sum of select unit mass
 235 resolution ions ($m/z > 147$) and does not represent the total siloxane fraction.
 236

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

243 HOMEChem and other indoor field studies provide insights to the complex processes that
 244 contribute to aerosol mass during cooking in real indoor environments.⁴ This work, in addition to
 245 prior reports^{8,27} highlights the potential for non-food and non-fuel related chemicals to contribute
 246 aerosol mass during indoor cooking and motivates future studies to consider the health effects of
 247 inhaling these compounds in airborne particles.
 248

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 257

258 Supplemental Information

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

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

264

265 **References**

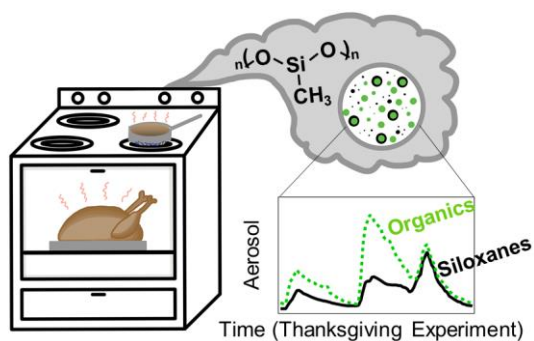
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