



1 **Contribution of Cooking Emissions to the Urban** 2 **Volatile Organic Compounds in Las Vegas, NV**

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21 **Abstract**

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23
24 Cooking is a source volatile organic compounds (VOCs) that degrades air quality. Cooking VOCs
25 have been investigated in laboratory and indoor studies, but the contribution of cooking to the
26 spatial and temporal variability of urban VOCs is uncertain. In this study, a proton-transfer-
27 reaction time-of-flight mass spectrometer (PTR-ToF-MS) is used to identify and quantify cooking
28 emission in Las Vegas, NV with supplemental data from Los Angeles, CA and Boulder, CO.
29 Mobile laboratory data show that long-chain aldehydes, such as octanal and nonanal, are
30 significantly enhanced in restaurant plumes and regionally enhanced in areas of Las Vegas with
31 high restaurant density. Correlation analyses show that long-chain fatty acids are also associated
32 with cooking emissions and the relative VOC enhancements observed in regions with dense
33 restaurant activity are very similar to the distribution of VOCs observed in laboratory cooking
34 studies. Positive matrix factorization (PMF) is used to quantify cooking emissions from ground
35 site measurements and compare the magnitude of cooking to other important urban sources, such
36 as volatile chemical products and fossil fuel emissions. PMF shows that cooking may account for
37 as much as 20% of the total anthropogenic VOC emissions observed by PTR-ToF-MS. In contrast,
38 emissions estimated from county-level inventories report that cooking accounts for less than 1%
39 of urban VOCs. Current emissions inventories do not fully account for the emission rates of long-



40 chain aldehydes reported here and further work is likely needed to improve model representations
41 of important aldehyde sources, such as commercial and residential cooking.

42

43 **1. Introduction**

44

45 Volatile organic compounds (VOCs) degrade air quality and are emitted to urban air from
46 many sources, including fossil fuel combustion (Warneke et al., 2012), the use of volatile chemical
47 products (VCPs, McDonald et al., 2018), industrial processes (Zhang et al., 2004), residential
48 heating and wood burning (e.g., McDonald et al., 2000; Coggon et al., 2016), cooking (Wernis et
49 al., 2022), and urban vegetation (e.g., Churkina et al., 2017). Each source emits a diverse set of
50 molecules that react alongside nitrogen oxides ($\text{NO} + \text{NO}_2 = \text{NO}_x$) to form ozone. Recent field
51 work in major US metropolitan areas has characterized the distribution of urban VOCs to assess
52 the chemical fingerprint of understudied emission sources, such as VCPs (Gkatzelis et al.,
53 2021b; Peng et al., 2022). These studies have shown that these sources emit oxygenated VOCs
54 (oVOCs) which react to form ozone and secondary organic aerosol. Models have been updated to
55 better describe the emissions and chemistry of select oVOCs, including alcohols, siloxanes,
56 glycols, and furanoids (Coggon et al., 2021; Pye et al., 2023; Qin et al., 2021).

57

58 Many oVOCs are emitted to urban air that have not been well-studied or incorporated into
59 air quality models (Karl et al., 2018). For example, McDonald et al. (2018) showed that C > 5
60 aldehydes measured in Los Angeles, CA could not be explained by emissions inventories that
61 contain VCPs, fossil fuels, or biogenic sources. Cooking is a source of oVOCs that is rich in
62 aldehydes and fatty acids (Klein et al., 2016a). Cooking VOCs have been extensively characterized
63 in the laboratory (Bastos and Pereira, 2010; Klein et al., 2016a; Klein et al., 2016b; Schauer et al.,
64 1999; Zhao and Zhao, 2018) and it has been shown that cooking is a key activity controlling the
65 budget of VOCs measured in indoor air (Arata et al., 2021; Klein et al., 2019; Klein et al., 2016a).
66 Numerous studies have shown that cooking is a ubiquitous and important component of organic
67 aerosol in urban areas (Hayes et al., 2013; Robinson et al., 2006; Robinson et al., 2018; Shah et al.,
68 2018; Slowik et al., 2010; Zhang et al., 2019) yet only a few studies have been conducted to
69 characterize cooking VOCs in ambient datasets (e.g., Peng et al., 2022; Wernis et al., 2022).

70

71 Cooking emissions result from the combustion and high temperature decomposition of
72 food and oils (Bastos and Pereira, 2010; Umamo and Shibamoto, 1987). During heating, fatty acids
73 undergo thermal oxidation to produce emissions of aldehydes, ketones, alcohols, acids, and other
74 products of depolymerization (Bastos and Pereira, 2010; Schauer et al., 1999). The use of spices
75 emits monoterpenes and their derivatives (Klein et al., 2016a). Studies that have speciated VOCs
76 from a variety of Western cooking styles (e.g., charbroiling, grilling, frying) and ingredients (e.g.,
77 oils, meats, and vegetables) show that aliphatic C₁-C₁₁ aldehydes account for a large fraction of
78 VOCs measured by gas-chromatography and mass spectrometry (e.g., Bastos and Pereira,
79 2010; Klein et al., 2016b; Peng et al., 2017; Schauer et al., 1999). For example, Klein et al. (2016b)



80 reported that aldehydes represent > 60% of the VOC mass emitted from frying or charbroiling
81 meats and vegetables.

82

83 Ambient observations have shown that long-chain aldehydes are present in urban air at
84 significant mixing ratios (e.g., nonanal ~ 100 – 200 ppt, Bowman et al., 2003; Wernis et al., 2022).
85 Recent studies have used hexanal and nonanal as markers for cooking emissions in Atlanta, GA
86 and Livermore, CA (Peng et al., 2022; Wernis et al., 2022). These species correlated with morning
87 and evening meal preparation, and it was suspected that restaurant emissions were a driving factor
88 for the observed temporal variability in Livermore (Wernis et al., 2022). In addition to cooking,
89 certain long-chain aldehydes are known to be emitted from diesel exhaust (e.g., hexanal, Gentner
90 et al., 2013), and some are produced from the emission and ozonolysis of oils and fatty acids
91 present in human skin, at the surface of ocean waters, or from other surfaces that contain
92 unsaturated lipids (Kruza et al., 2017; Liu et al., 2021; Wang et al., 2022; Kilgour et al., 2021).

93

94 The high abundance of aliphatic aldehydes from cooking suggests that they may be useful
95 markers to constrain cooking VOC emissions in urban areas. The utility of aldehydes as cooking
96 markers relies on the characterization of their sources in the atmosphere as well as careful
97 characterization of measurement techniques used to detect these species. Short-chain aldehydes
98 ($C < 5$) are unlikely to serve as useful markers because they are produced in the atmosphere from
99 the OH oxidation of primary organic molecules and are also directly emitted from fossil fuel
100 emissions and biomass burning (Gentner et al., 2013; Koss et al., 2018; de Gouw et al., 2018). Long-
101 chain aliphatic aldehydes ($C > 6$) have the potential to be useful markers for cooking in urban
102 areas, but their primary sources, spatial distributions, and abundances in the atmosphere remain
103 uncertain. Some long-chain aldehydes may be emitted from mobile sources (e.g., Gentner et al.,
104 2013) while others may be emitted from surface ozone chemistry (e.g., Liu et al., 2021). No field
105 measurements have reported the spatial distribution of long-chain aldehydes to determine likely
106 sources in urban air.

107

108 This study evaluates the contribution of commercial and residential cooking emissions to
109 urban VOCs using mobile laboratory and ground site observations in Las Vegas, NV with
110 supplemental observations made Los Angeles, CA and Boulder, CO. Mobile laboratory
111 measurements show that long-chain aliphatic aldehydes are significantly enhanced downwind of
112 restaurants and exhibit spatial distributions in urban regions of Las Vegas that closely matches
113 restaurant density. Furthermore, these measurements show that the distribution of VOCs that
114 correlate with long-chain aldehydes strongly resembles the distribution observed in restaurant
115 plumes. We conduct a source apportionment analysis to determine the extent to which cooking
116 emissions impact urban VOCs relative to other important anthropogenic sources, such as motor
117 vehicles and VCPs, and compare these observations to commonly used emissions inventories.

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120 **2. Methods**

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122 **2.1. Field Campaign Description**

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124 Air quality measurements were performed during the 2021 Southwest Urban NO_x and VOC
125 Experiment (SUNVEx) and Re-Evaluating the Chemistry of Air Pollutants in California (RECAP-
126 CA) studies. Trace gases were measured from the NOAA mobile laboratory and at ground sites
127 located at the Jerome Mack Air Quality Station in Las Vegas, NV and the California Institute of
128 Technology campus in Pasadena, CA. In Las Vegas, seven mobile laboratory drives were
129 conducted between 27 June and 31 July, 2021 with a focus on sampling densely populated regions.
130 Ground sampling was primarily conducted from 30 June– 27 July, 2021.

131

132 Figure 1 is a map of the Las Vegas valley that shows the locations of residential, commercial,
133 and entertainment districts. The blue dots show the locations of restaurants that are cataloged in
134 health inspection reports maintained by the Southern Nevada Health District (SNHD, 2022). The
135 wind rose is centered at the Jerome Mack Air Quality Station and shows the prevailing wind
136 directions and speeds observed at the ground site. The mobile laboratory sampled across the region
137 (e.g., Figure 3), but focused drives was conducted along the Las Vegas Strip (the purple shaded
138 region shown in Fig. 1). The Las Vegas Strip is an entertainment district with a high density of
139 casinos, hotels, bars, and restaurants that emit VCP, fossil fuel, and cooking VOCs. The Jerome
140 Mack ground site is located ~8km northeast of the Las Vegas Strip in a residential area with
141 restaurants and small commercial businesses located along major streets. The prevailing wind
142 patterns show that ground measurements were routinely impacted by air transported from the Las
143 Vegas Strip along with regions to the north with higher commercial activity.

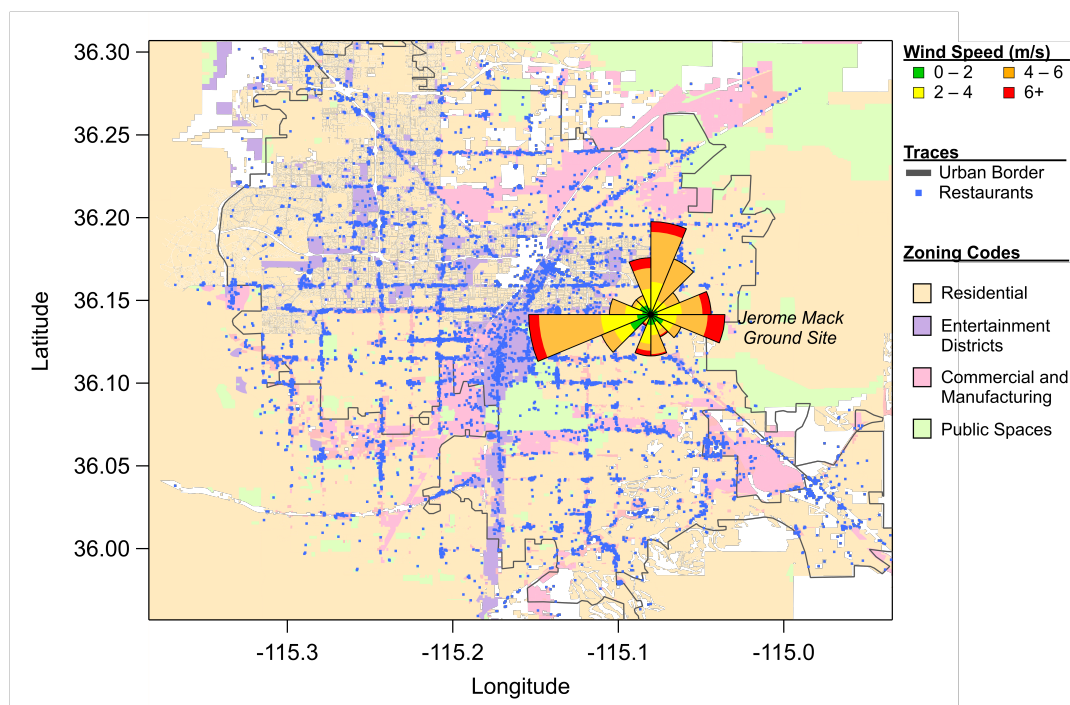
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145 Ground measurements during RECAP-CA were performed at the California Institute of
146 Technology in Pasadena, CA from 1 August – 5 September, 2021. These measurements serve as
147 a comparison to the observations in Las Vegas. VOCs and other trace gases were sampled from
148 the top of a 10 m tower. The location of the ground site was located within 0.5km from the ground
149 site used during the 2010 CalNex campaign (Ryerson et al., 2013). The Pasadena ground site has
150 been previously characterized and is a downwind receptor site for air impacted by emissions from
151 downtown Los Angeles, CA (e.g., de Gouw et al., 2018).

152

153 Supplemental mobile laboratory measurements were performed in Los Angeles, CA and
154 Boulder, CO to sample VOCs downwind of individual restaurants. These measurements serve as
155 comparisons against the restaurant emissions observed in the Las Vegas region.

156



157

158 **Figure 1:** Zoning map of the Las Vegas region showing areas permitted for residential buildings,
159 entertainment districts, commercial and manufacturing use, and public spaces. Data are available from the
160 Clark County GIS Management Office (<https://clarkcountygis-cgismo.hub.arcgis.com>, accessed
161 September 2023) The blue dots show the locations of restaurants in the Las Vegas valley that are cataloged
162 in health inspection reports maintained by the Southern Nevada Health District (SNHD, 2022). The wind
163 rose shows the prevailing wind directions and speed observed at the Jerome Mack ground site. The wind
164 rose is centered on the map at the location of Jerome Mack.

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166 2.2. Instrument Description

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168 PTR-ToF-MS

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170 VOC mixing ratios were measured using a Vocus proton-transfer-reaction time-of-flight mass
171 spectrometer (PTR-ToF-MS) (Yuan et al., 2016;Krechmer et al., 2018). The PTR-ToF-MS
172 measures a large range of aromatics, alkenes, nitrogen-containing species, and oxygenated VOCs.
173 A full description of the instrument during SUNVEX is provided by Coggon et al. (2023). Briefly,
174 the PTR-ToF-MS sampled air a $\sim 2 \text{ L min}^{-1}$ through a short ($< 1\text{m}$) inlet while in the mobile
175 laboratory. At the ground sites, the instrument sampled at $\sim 20 \text{ L min}^{-1}$ from a 10 m tower. The
176 Vocus drift tube was operated at 110°C with an electrical field (E) to number density (N) ratio
177 (E/N) of 140 Td. Instrument backgrounds were determined every 2 h for ground site experiments
178 and every $\sim 15\text{-}30$ minutes during drives by passing air through a platinum catalyst heated to 350°C .



179 Data were processed following the recommendations of Stark et al. (2015) using the Tofware
180 package in Igor Pro (WaveMetrics). The PTR-ToF-MS was calibrated using gravimetrically-
181 prepared gas standards for typical VOCs such as acetone, methyl ethyl ketone, toluene, and C8-
182 aromatics. Many compounds unavailable in gas standards were quantified by liquid calibration
183 methods as described by Coggon et al. (2018). This included D5-siloxane,
184 parachlorobenzotriflouride, octanal and nonanal. All other compounds were quantified using
185 estimated proton-transfer-reaction rate constants as described by Sekimoto et al. (2017). Further
186 corrections were applied to masses assigned to long-chain aldehydes based on observed mass-
187 dependent changes in fragmentation patterns described in the Supplemental Information and
188 shown in Fig. S2.

189

190 PTR-ToF-MS only resolves VOC molecular formula. To identify structural isomers, a
191 custom-built gas-chromatography (GC) instrument was used to collect and pre-separate VOCs
192 prior to detection by PTR-ToF-MS. A full description of the system is provided by Stockwell et
193 al. (2021) and its operation during SUNVEx is described by Coggon et al. (2023). Briefly, the GC
194 consists of a DB-624 column (Agilent Technologies, 30 m, 0.25 mm ID, 1.4 μm film thickness)
195 and oven identical to the system described by Lerner et al. (2017). VOCs were condensed onto a
196 liquid nitrogen cryotrap, flash vaporized, then passed through the column using nitrogen as a
197 carrier gas. The column was linearly heated during separation from 40-150°C. The effluent from
198 the column was directly injected in the PTR-ToF-MS inlet. At the Jerome Mack ground site, GC
199 samples were collected every 2 hours and immediately analyzed by PTR-ToF-MS. GC samples
200 were also collected during a nighttime mobile laboratory drive on July 31, 2021. These samples
201 were used to help interpret PTR-ToF-MS measurements along the Las Vegas Strip.

202

203 A key goal of this study is to characterize the spatial and temporal pattern of long-chain
204 aldehydes. Aldehydes and ketone isomers are quantified by PTR-ToF-MS using measurements of
205 the proton-transfer product ions (= VOC mass + H^+). Aliphatic aldehydes also undergo
206 dehydration (= VOC mass + H^+ - H_2O) and fragmentation reactions, which effectively lowers the
207 instrument sensitivity to the proton-transfer product. Consequently, it can be challenging to
208 unambiguously assign carbonyl ions to specific isomers. In the Supplemental Information, GC-
209 PTR-ToF-MS and mobile laboratory PTR-ToF-MS measurements show that C_8 and C_9 carbonyls
210 measured in urban areas are predominantly associated with octanal (detected at m/z 129,
211 $\text{C}_8\text{H}_{16}\text{OH}^+$) and nonanal (detected at m/z 143, $\text{C}_9\text{H}_{18}\text{OH}^+$). These ions have no detectable
212 interferences from ketone isomers in GC-PTR-ToF-MS spectra (Fig. S1) and the ratio of these
213 ions with carbonyl dehydration products most closely matches the fragmentation patterns of
214 aldehydes (Fig. S3). Smaller carbonyls have significant interferences from ketone isomers, which
215 complicates their use as markers for aldehyde emissions. Here, we focus on the spatial and
216 temporal trends of octanal and nonanal and use these markers to determine the fingerprint of
217 cooking VOC emissions.

218



219 **LGR Carbon Monoxide**

220

221 Carbon monoxide was measured at the Caltech ground site from a 10 m stainless steel tube
222 (3.2 mm OD, 1.6 mm ID) using off-axis integrated cavity output spectroscopy (ABB Inc./Los
223 Gatos Research model F-N₂O/CO-23r) (Roberts et al., 2022). Data were measured at 1-Hz and
224 reported as 1-minute averages. Instrument precision was estimated to be ± 0.2 ppb ($1-\sigma$), and the
225 $1-\sigma$ uncertainty was estimated to be $\pm 1\%$ based on calibrations in the laboratory before and after
226 the SUNVEx/RECAP-CA project.

227

228 **2.3. Positive Matrix Factorization**

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230 Positive matrix factorization (PMF) is used to analyze the PTR-ToF-MS data and apportion
231 VOCs to cooking and other urban sources in Las Vegas. PMF was conducted using the Source
232 Finder (SoFi) software package in Igor Pro (Canonaco et al., 2013). Two periods are analyzed
233 when PTR-ToF-MS measurements were available: 30 June–9 July and 19–27 July. In this analysis,
234 we constrain PMF with a mobile source profile derived from mobile measurements following the
235 recommendations of Gkatzelis et al. (2021b). We present a solution of factors representing mobile
236 sources, VCPs, cooking, and regional chemical oxidation. A full description of the PMF analysis
237 is provided in the Supplemental Information.

238

239 **3. Results**

240

241 **3.1. Long-chain aldehydes downwind of restaurants**

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243 Previous studies have shown that cooking organic aerosols (COA) from dense restaurant
244 clusters exhibit plume-like behavior that can impact local air quality at spatial scales of 0.5–1 km
245 (Robinson et al., 2018; Shah et al., 2018). For example, Robinson et al. (2018) showed that organic
246 aerosol was enhanced by as much as 100–200 $\mu\text{g m}^{-3}$ within 1 km of restaurants and resulted in
247 average local organic aerosol enhancements of 3.2 $\mu\text{g m}^{-3}$. Consequently, it is expected that VOCs
248 would also be significantly enhanced in close proximity of restaurants.

249

250 Figure 2 shows mobile laboratory measurements of nonanal and octanal mixing ratios
251 downwind of two fast food restaurants in Los Angeles, CA and Boulder, CO. Mixing ratios of
252 markers typically representative of personal care products (D5-siloxane) and motor vehicle
253 emissions (benzene) are also shown to highlight the presence of other sources in the region. The
254 restaurant in Los Angeles primarily serves hot dogs, while the restaurant in Boulder serves
255 hamburgers and fried foods. The highlighted boxes show periods where the mobile laboratory was
256 parked to sample restaurant emissions. All other data reflect sampling periods when the mobile
257 laboratory was driven through densely populated areas of Los Angeles and Boulder. In both cases,
258 the mobile laboratory was parked within 50m of the restaurant exhausts.



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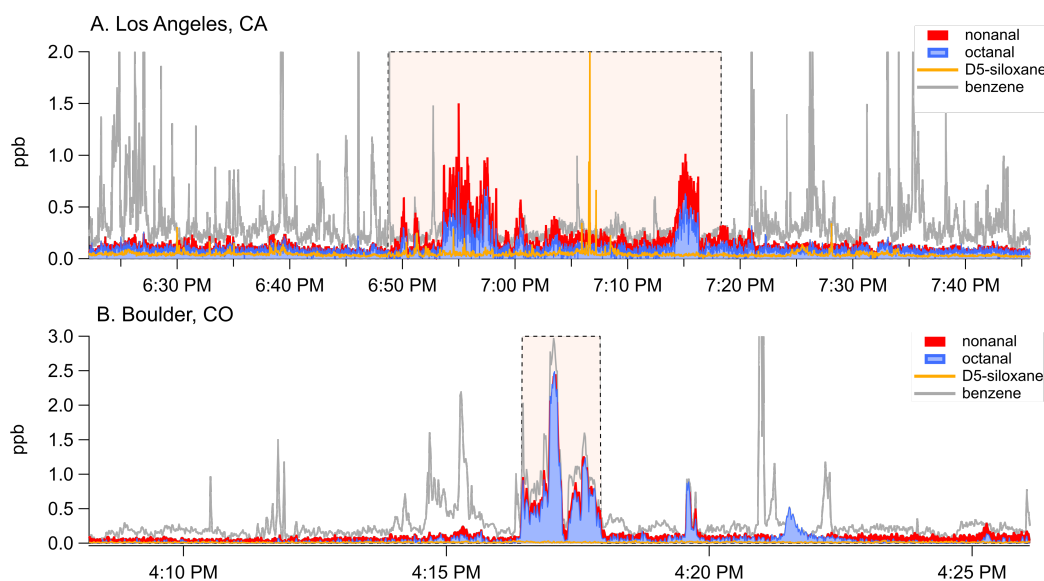
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Aldehyde mixing ratios downwind of both restaurants exceeded 1 ppb. Generally, these mixing ratios were elevated relative to the surrounding densely populated regions. Octanal and nonanal mixing ratios were not significantly enhanced in tailpipe emissions, which is consistent with previous studies showing that on-road emission factors of these from US vehicles are low (Gentner et al., 2013). Octanal and nonanal were not strongly correlated with mixing ratios of D5-siloxane, though there were periods when long-chain aldehyde and D5-siloxane enhancements were coincident. This may result from the co-location of food and people or a possible human emission source. Octanal and nonanal are known to be produced from skin ozonolysis (Liu et al., 2021; Wang et al., 2022) and carbonyls are potential ingredients in fragranced consumer products, though emissions inventories and measurements of fragrance formulations do not indicate that octanal and nonanal are significant ingredients of VCPs (Hurley et al., 2021; McDonald et al., 2018; Yeoman et al., 2020).



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Figure 2: Mobile laboratory measurements of octanal, nonanal, D5-siloxane, and benzene in (A) Los Angeles, CA and (B) Boulder, CO. The shaded regions show periods when the mobile laboratory was parked downwind of a restaurant that primarily serves hot dogs (Los Angeles) and a restaurant that primarily serves hamburgers (Boulder). All other data were collected while the mobile laboratory sampled air in populated areas.

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Figure 2 shows that restaurants are a strong source of aliphatic aldehydes, such as octanal and nonanal. Based on these enhancements, it is likely that VOCs emitted from cooking are significantly enhanced in regions with dense restaurant activity. These inferences are consistent with previous mobile laboratory observations of primary organic aerosols. For example, Robinson

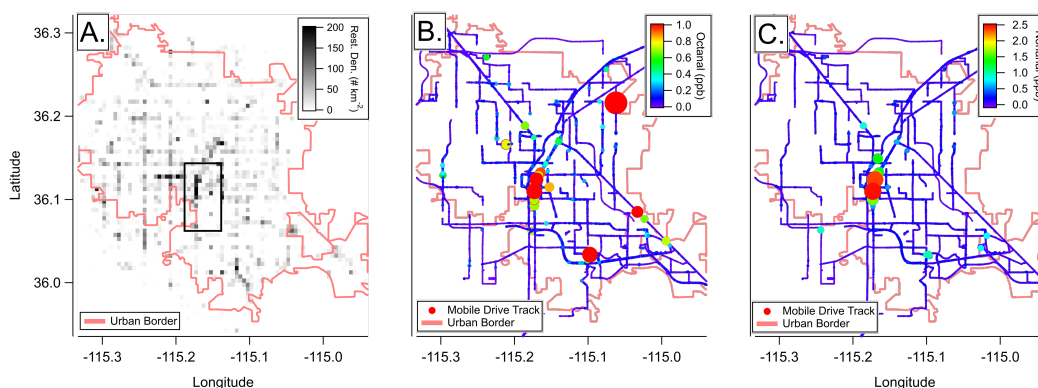


284 et al. (2018) found that organic aerosol in commercial districts of Pittsburgh, PA with significant
285 restaurant density was nearly twice as concentrated as organic aerosol in areas with highways and
286 significant traffic. Similar results were observed by Shah et al. (2018) in Oakland, CA where COA
287 constituted ~ 50% of the primary organic aerosol and was observed to be enhanced in downtown
288 regions where restaurant density was highest. Both studies demonstrate that air quality in urban
289 areas is significantly impacted by restaurant density.

290

291 Las Vegas is a sprawling city where most emission sources are concentrated along the Las
292 Vegas Strip (Figure 1). Figure 3 shows the spatial distribution of octanal and nonanal from all of
293 the mobile laboratory drives, along with a map of restaurant density calculated using the restaurant
294 location data shown in Figure 1. Each pixel is determined by summing the number of restaurants
295 over a 0.5 x 0.5 km grid. Figure 3 shows that octanal and nonanal are well-correlated ($R^2 = 0.82$)
296 and predominantly enhanced in the Las Vegas Strip area where anthropogenic emissions are the
297 highest. Figure 3A shows that this region has a high restaurant density compared to other regions
298 of the Las Vegas Valley. We note that brief (~1s), isolated enhancements in PTR-ToF-MS
299 measurements of octanal are observed outside of the Las Vegas Strip. These enhancements also
300 have corresponding increases in nonanal, though the ratios of these species are different than what
301 is observed along the Las Vegas Strip.

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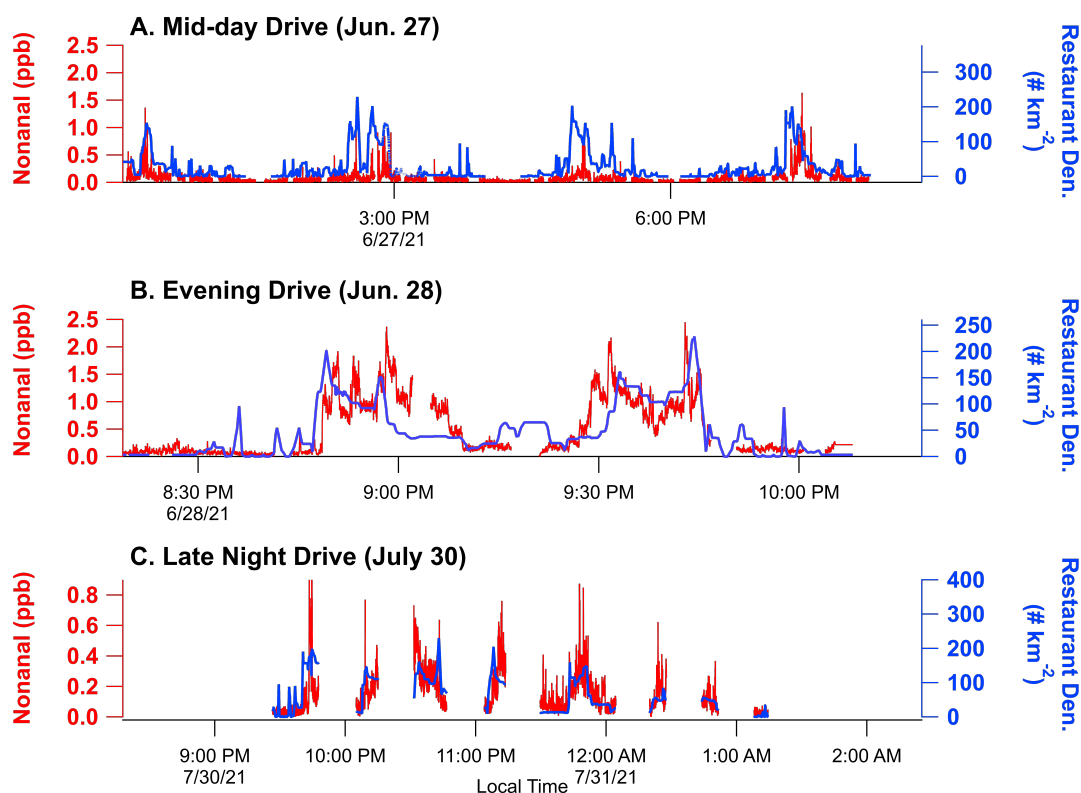
305 **Figure 3:** (A) Restaurant density in Clark County, NV. Restaurant density is determined using the
306 restaurant locations shown in Fig. 1 and is calculated by summing the number of restaurants located within
307 a 0.5 x 0.5 km grid. The rectangle shows the approximate region of the Las Vegas Strip. Panels (B) and (C)
308 show the mobile laboratory path colored by octanal and nonanal mixing ratios, respectively. Markers are
309 sized by the corresponding mixing ratios shown in the color scales.

310

311 Figure 4 further evaluates the spatial distribution of long-chain aldehydes by comparing
312 nonanal mixing ratios against the restaurant density in the proximity of the mobile laboratory.
313 Here, restaurant density is extracted from Fig. 3A by selecting the grid cells that are coincident
314 with the mobile laboratory position while sampling in the vicinity of the Las Vegas Strip (indicated



315 by the black box shown in Fig. 3A). The three drives shown correspond to mid-day (12:00 – 7:00
316 PM local time), evening (8:00 – 11:00 PM), and late-night (9:30 PM – 1:00 AM) sampling. The
317 drives show that nonanal is generally enhanced in regions with higher restaurant density. Nonanal
318 mixing ratios are highest during evening drives when activities from the entertainment industry in
319 the Las Vegas Strip area, including dining, are likely most frequent (panels B and C).
320 Enhancements in nonanal are also observed during a mid-day drive (panel A), though mixing ratios
321 are generally lower due to a higher boundary layer and potentially lower emissions in the Las
322 Vegas Strip area during the day. Octanal exhibits a similar relationship with restaurant density.
323 These results are consistent with the observed enhancements in organic aerosol seen previously
324 observed in dense restaurant regions (Robinson et al., 2018; Shah et al., 2018).
325



326 **Figure 4:** Nonanal mixing ratios and corresponding restaurant density in areas sampled by the mobile
327 laboratory. Restaurant density is determined by averaging restaurant data in Figure 3A on a 0.5 km x 0.5
328 km grid, then extracting data along the mobile laboratory drive track.
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332 Figure S12 shows that the octanal and nonanal observed along the Las Vegas Strip are also
333 measured at the Jerome Mack ground site. The two species are well-correlated ($R^2 > 0.86$) and
334 most abundant at night, likely due to a combination of meteorology (i.e., shallow nocturnal



335 boundary layer) and higher emissions in the evening (e.g., Fig. 4). Figure S12 also shows octanal
336 and nonanal mixing ratios observed at the Caltech ground site in Pasadena, CA. These
337 measurements exhibit similar temporal behavior and demonstrate that long-chain aldehyde are
338 ubiquitous in many urban regions. We note that nonanal ratios reported here are similar to those
339 observed from previous studies (~ 100 – 200 ppt, Bowman et al., 2003;Wernis et al., 2022).

341 **3.2. Species co-emitted with octanal and nonanal in the Las Vegas Strip**

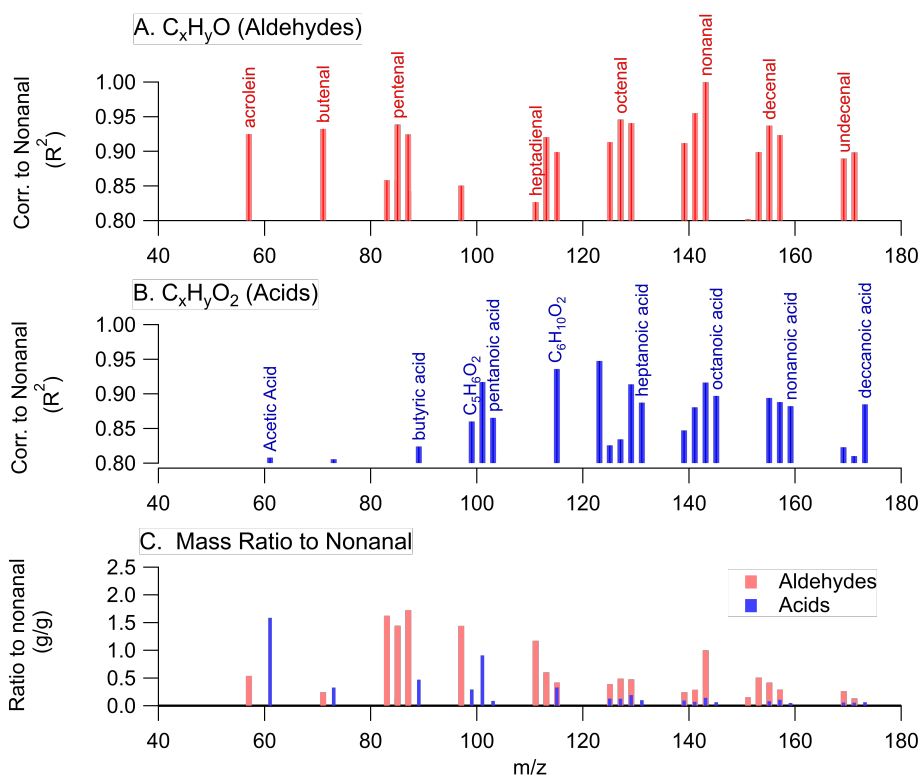
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343 Octanal and nonanal represent a significant fraction of cooking VOCs measured in laboratory
344 studies (>5%) but are just two of many VOCs emitted from cooking activities (Klein et al., 2016b).
345 To assess the potential fingerprint of VOCs in regions impacted by commercial cooking, we
346 evaluate the VOC mass spectra from the Las Vegas Strip and identify cooking VOCs by correlating
347 PTR-ToF-MS ions to observations of nonanal. Species are only included in this analysis if the
348 detected ion likely represents a proton-transfer product (i.e., fragments are excluded) and correlates
349 with nonanal with $R^2 > 0.8$. This high correlation coefficient is used to identify potential co-emitted
350 species, while excluding VOCs emitted from sources that are co-located with restaurants in the
351 Las Vegas Strip, such as mobile sources and VCPs. For example, D5-siloxane correlates with
352 nonanal with an R^2 of 0.78. Personal care product emissions are significant along the Las Vegas
353 Strip (D5-siloxane mixing ratios > 1 ppb on June 28, 2021), but octanal and nonanal have not been
354 reported as major components of fragranced personal care products (e.g., McDonald et al.,
355 2018;Steinemann, 2015;Steinemann et al., 2011;Yeoman et al., 2020;Hurley et al., 2021). A high
356 correlation is expected because food and people are co-located; however, in this analysis D5-
357 siloxane and compounds with $R^2 < 0.8$ are excluded.

358
359 Figure 5 shows the correlation spectrum of the VOCs to nonanal. The correlation coefficient
360 and the ratio of VOCs to nonanal are plotted versus the detected mass. The compounds that
361 correlate with nonanal have chemical formula of either C_xH_yO or $C_xH_yO_2$ with carbon numbers
362 ranging between $C_2 - C_{11}$. Compounds with formula C_xH_yO are likely aliphatic aldehydes with
363 varying degrees of saturation and some key species are highlighted for each carbon grouping
364 (Figure 6A). Species with the highest correlation to nonanal include octenal ($R^2 = 0.94$), decenal
365 ($R^2 = 0.93$), butenal/crotonaldehyde ($R^2 = 0.93$), and acrolein ($R^2 = 0.92$). While individual $C_3 -$
366 C_5 species are the largest contributors to the total signal, the sum of long-chain aldehydes ($C_6 -$
367 C_{11}) are >50% of the total spectrum.

368
369 Compounds with formula $C_xH_yO_2$ likely correspond to fatty acids (Figure 5B). Gaseous acids
370 are released from the high temperature decomposition of long-chain acids present in meat and
371 previous studies have measured significant emissions of heptanoic, octanoic, nonanoic, and
372 decanoic acid (Schauer et al., 1999;Klein et al., 2016b). The acids in Figure 5B are some of the
373 most abundant acids observed along the Las Vegas Strip. The strong correlation of nonanal to fatty
374 acids further supports that cooking emissions are an important emitter of long-chain aldehydes in
375 this region.



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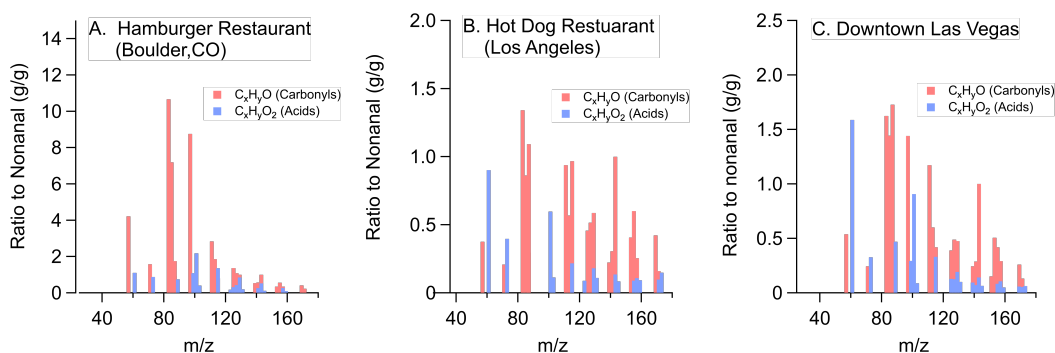
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Figure 5: VOC correlation to nonanal in downtown Las Vegas during nighttime mobile sampling for (A) C_xH_yO species (assigned as aldehydes) and (B) C_xH_yO₂ species (assigned as acids). (C) VOC / nonanal ratios for species that correlate with nonanal with R² > 0.8.

Figure 6 compares the spectra measured along the Las Vegas Strip to the spectra derived from the individual restaurants sampled in Boulder, CO and Los Angeles, CA (Figure 2). The VOC ratios observed in the restaurant plumes are strikingly similar to the ratios observed in downtown Las Vegas. The Las Vegas samples most resemble those observed downwind of the hot dog restaurant in Los Angeles. The two spectra are well correlated (R² = 0.82) which further supports that the aldehydes and acids measured along the Las Vegas Strip were associated with restaurant emissions. The spectra are also comparable to available PTR-ToF-MS spectra of meat cooking emissions reported by Klein et al. (2016b). These laboratory measurements show that heptadienal, octenal, nonanal, decadienal, and undecanal are key C₇ – C₁₁ aldehydes emitted when cooking meats with vegetable oils. The same aldehydes are observed in the Las Vegas Strip area.



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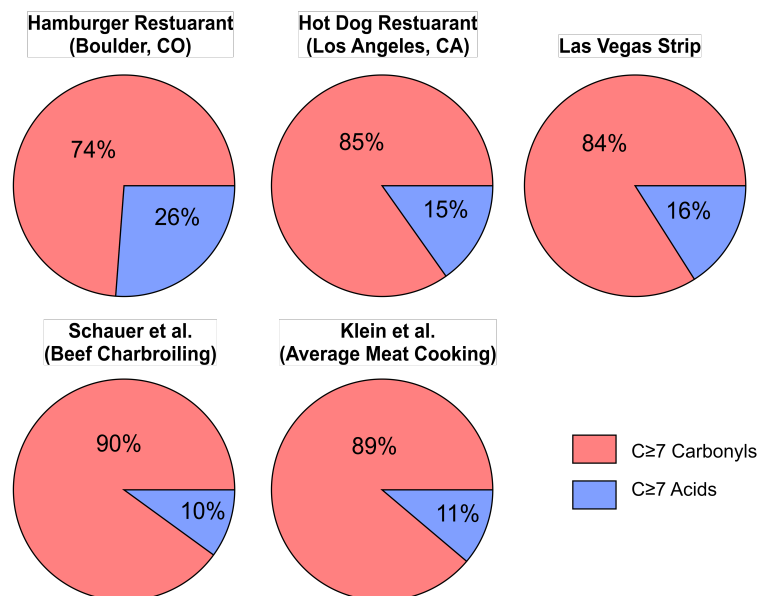
395 **Figure 6.** Comparison of (A) restaurant emissions from a hamburger restaurant in Boulder, CO, (B)
396 restaurant emissions from a hot dog restaurant in Los Angeles, and (C) cooking emissions from
397 measurements along the Las Vegas Strip.

398

399 Klein et al. (2016b) speciated laboratory cooking emissions and distinguished emissions of
400 higher carbon aldehydes ($C \geq 7$) and acids from lower carbon species. These groupings are also
401 distinct in the laboratory VOC distributions reported by Schauer et al. (1999) and are suspected to
402 be signatures of cooking emissions in Las Vegas (e.g., Fig. 5). Figure 7 compares the distribution
403 of $C \geq 7$ oxygenates observed in this study (top row) with the distributions reported by laboratory
404 studies (bottom row). Schauer et al. (1999) observed that ~10% of the $C \geq 7$ mass emitted from
405 beef charbroiling was attributed to acids, while 90% was attributable to carbonyls largely
406 composed of aldehydes. Klein et al. (2016b) observed a similar profile averaged across a range of
407 experiments using meats and vegetable oils. In the Las Vegas Strip area, acids represented ~16%
408 of mass associated with $C \geq 7$ compounds, while the remainder is associated with carbonyls
409 dominated by aldehydes. Carbonyls are the dominant $C \geq 7$ emissions from both restaurants
410 sampled by the mobile laboratory. The similarity in the distributions between laboratory and field
411 observations further suggest that long-chain aldehydes are useful markers for constraining cooking
412 emissions in urban air.

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415

416 **Figure 7:** Comparison of the $C \geq 7$ oxygenates measured as part of this study (top row) with those observed
417 from laboratory experiments reported by Schauer et al. (1999) and Klein et al. (2016b) (bottom row). The
418 distribution from Schauer et al. (1999) reflects emissions from beef charbroiling, while the distribution
419 from Klein et al. (2016b) is derived as the average distribution from the frying of pork, chicken, beef, and
420 fish in a range of vegetable oils.

421

422 **4. Source apportionment from Las Vegas measurements**

423

424 The analysis described in Section 3.2 provides a perspective of the key VOCs emitted from
425 commercial cooking. Other VOCs are also likely associated with cooking but are not resolved by
426 a simple correlation analysis due to the presence of other important sources along the Las Vegas
427 Strip, such as ethanol from VCPs or monoterpenes from fragranced consumer products. Moreover,
428 emissions from residential cooking may also contribute to regional VOC mixing ratios. Here, we
429 discuss the PMF results to determine the emissions profile and the contribution of cooking to total
430 VOC emissions observed at the Jerome Mack ground site.

431

432 Figures 8 and 9 show the PMF solution for the data collected at the Jerome Mack ground site.
433 Figure 8 shows the time series and factor profiles, while Fig. 9 shows the average diurnal profiles.
434 We present a 5-factor solution where VOCs are apportioned to (1) a mobile source factor, (2) a
435 VCP-dominated factor, (3) a cooking-dominated factor, (4) a regional background plus secondary
436 oxidation processes, and (5) a local solvent source. A full description of the PMF results is
437 provided in the Supplemental Information.

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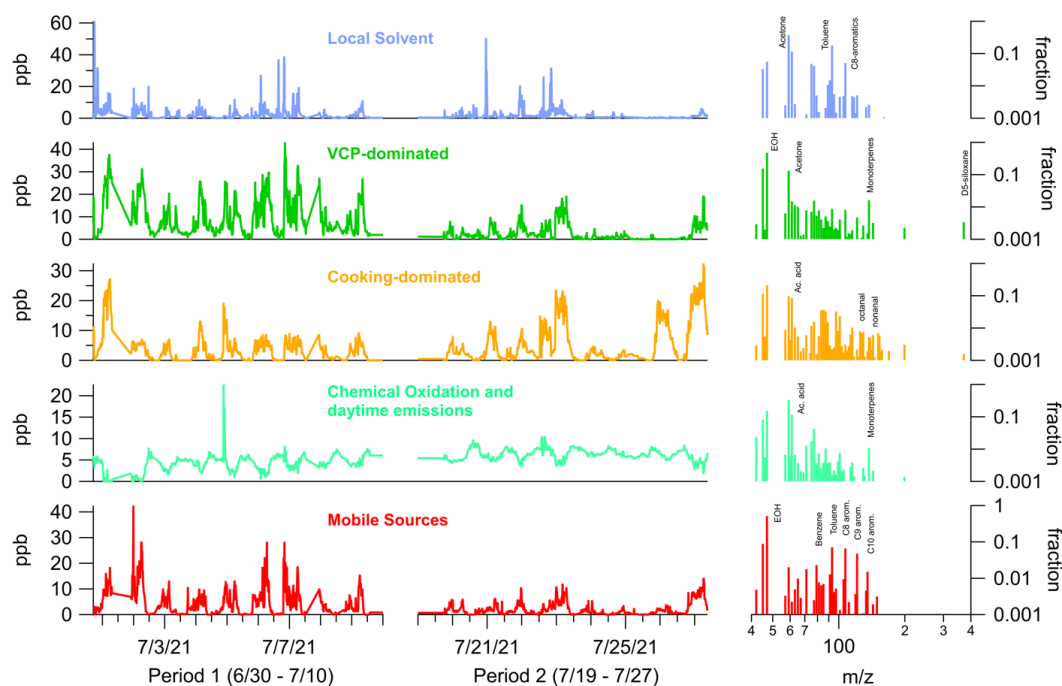


439 The mobile source factor is largely composed of ethanol and C₆-C₁₀ aromatics (Fig. S4). The
440 VCP-dominated factor is primarily composed of ethanol (EOH), but also contains D5-siloxane,
441 monoterpenes, and acetone, which are common ingredients in consumer products. Both factors
442 resemble the solution presented by Gkatzelis et al. (2021b) for New York City. The VCP-
443 dominated factor derived here has two key differences from the factor derived by Gkatzelis et al.
444 (2021b). First, the VCP-dominated factor from New York City contained a series of other VCP
445 markers, including PCBTF and D4-siloxane. These molecules are largely associated with
446 construction activity and are expected to be emitted from the application of industrial coatings and
447 adhesives (Gkatzelis et al., 2021a; Stockwell et al., 2021). At the Jerome Mack ground site, PCBTF
448 variability was largely attributed to the local solvent factor (Fig. 8), which appeared to come from
449 a point source located near the ground site. Second, the VCP-dominated factor reported by
450 Gkatzelis et al. (2021b) also contained methyl ethyl ketone, which is a prominent solvent in
451 consumer and industrial VCPs. Methyl ethyl ketone was excluded from this analysis due to a water
452 cluster interference produced within the Vocus drift tube.

453

454 The oxidation factor is largely composed of multiple oxygenated carbon-containing masses,
455 which agrees with secondary factors resolved by PMF in other cities (Gkatzelis et al., 2021b; Peng
456 et al., 2022). At the Jerome Mack site, this factor also contained VOCs that are emitted during
457 daytime hours from biogenic sources (e.g., isoprene, monoterpenes) due to emissions from urban
458 vegetation. In general, isoprene and monoterpenes had relatively low mixing ratios over the
459 analyzed sampling period (< 150 ppt). For comparison, measurements in Los Angeles during
460 RECAP-CA show that average isoprene mixing ratios exceeded 2 ppb (Coggon et al., 2023). This
461 difference highlights that urban vegetation emissions in Las Vegas are significantly lower than
462 other cities.

463



464
 465 **Figure 8:** 5-factor PMF solution for the ground site data at Jerome Mack. Shown are the factor time profiles
 466 for the two time periods considered for this analysis (Period 1, 30 June–10 July and Period 2, 19–27 July)
 467 along with the resolved factor profiles.
 468

469 PMF of the Jerome Mack data resolves a factor that is enriched in aldehydes, which we
 470 interpret as the cooking-dominated factor. This factor includes contributions from octanal,
 471 nonanal, acetic acid, acrolein, and higher carbon aldehydes and acids, which is consistent with the
 472 cooking emissions observed along the Las Vegas Strip and downwind of restaurants. Figure S10
 473 compares the PMF profile to the VOC/nonanal profiles resolved by the mobile laboratory and
 474 shows that the two profiles agree for overlapping species. This agreement supports the PMF
 475 resolution of mass associated with important cooking VOCs. The PMF factor also includes
 476 ethanol, monoterpenes, and acetone/propanal, which were not resolved by the mobile laboratory
 477 analysis. Cooking emits significant amounts of ethanol and monoterpenes to indoor air (Arata et
 478 al., 2021; Klein et al., 2016a), and is a dominant source of total VOC emissions in residential indoor
 479 air (Arata et al., 2021; Klein et al., 2019). These species represent ~22% of the cooking-dominated
 480 factor.
 481

482 Figure 9 shows daily mass concentrations for each factor. Gkatzelis et al. (2021b) compared
 483 the speciation profiles of the VCP-dominated and mobile source profiles to those represented in
 484 emissions inventories and found that ~53% of the mass associated with mobile source emissions
 485 and ~50% of the mass associated with VCPs results from emissions that cannot be resolved by

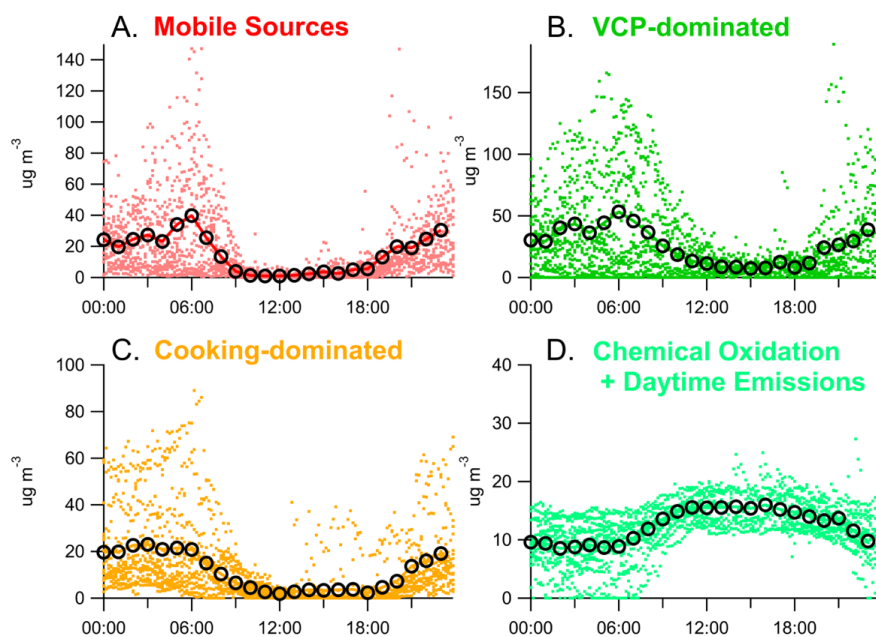


486 PTR-ToF-MS (e.g., alkanes and small alkenes). The mobile source and VCP-dominated factors
487 shown in Figures 9 and 10 have been adjusted to account for this unresolved mass. No adjustments
488 are applied to the cooking-dominated, solvent, or oxidation factor as it is assumed that PTR-ToF-
489 MS measures the key VOCs from these sources. This may not account for mass that has been
490 previously reported from cooking emissions, such as alkanes or alkenes (Schauer et al., 1999).

491

492 For the mobile source, VCP-dominated, and cooking-dominated factors, mass concentrations
493 are highest at night when the nocturnal boundary layer is shallow. In the daytime, the boundary
494 layer rapidly rises to as high as 4 km (Langford et al., 2022), and VOC concentrations decrease in
495 response. In contrast, the chemical oxidation + daytime emissions factor increases during daytime
496 hours, further supporting that this factor is largely driven by secondary processes.

497



498

499 **Figure 9:** Diurnal patterns for the four factors resolved by PMF at the Jerome Mack ground site. The black
500 circles show the hourly mean values calculated over the full PMF solution.

501

502 To normalize for the impact of meteorology, Figure 10A shows the fraction that each primary
503 factor contributes total VOC mass resolved by PMF at the Jerome Mack ground site. Here, total
504 anthropogenic emissions are represented as the sum of the VCP-dominated, mobile source, and
505 cooking-dominated factors. The local solvent factor is excluded from this analysis since it is not
506 representative of regional VOC concentrations. Figure 10B shows the average PMF solution over
507 the entire analysis period.



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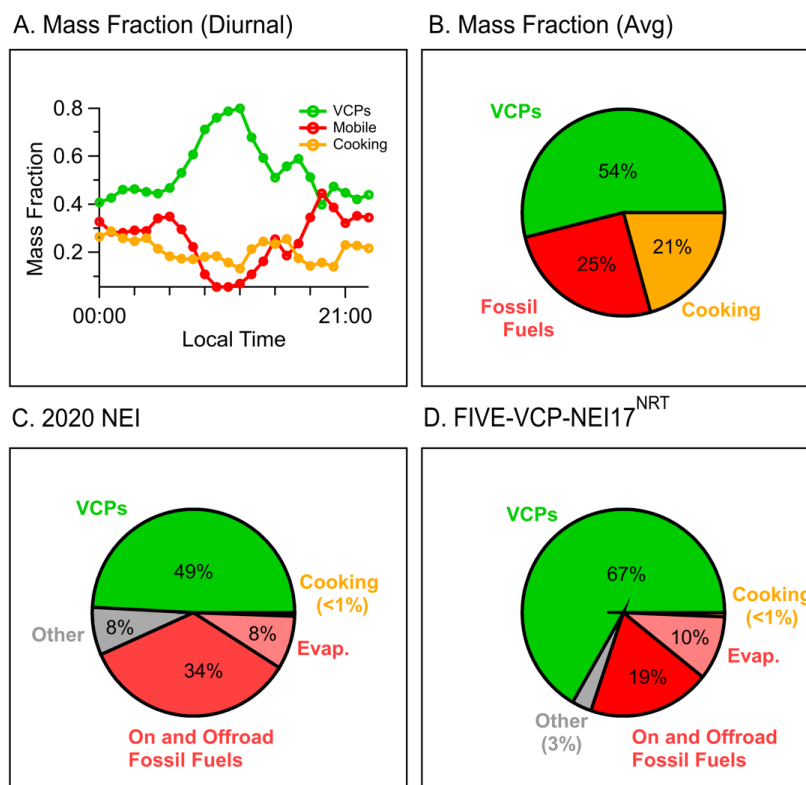
509 Figure 10A shows that each factor contributes to the total anthropogenic emissions at different
510 times of day depending on the emission patterns. The VCP-dominated factor is the largest
511 contributor to total VOC emissions in Las Vegas and constitutes 40 – 80% of the primary VOCs
512 resolved by PMF. These concentrations are largely driven by the high emissions of solvents, such
513 as ethanol and acetone, which is consistent with observations from NYC (Gkatzelis et al., 2021b).
514 VCP emissions exhibit the highest relative abundances early in the day (~11:00 AM), then
515 decrease in relative abundance throughout the day. This behavior is similar to the diurnal pattern
516 of personal care product emissions observed in cities such as Boulder, CO where the mixing ratios
517 of D5-siloxane from deodorants and hair products peak during morning hours and decayed as
518 personal care products evaporate (Coggon et al., 2018). During evening and rush hour periods,
519 mobile sources constitute ~ 30-40% of the total primary VOC mixing ratios, but then decrease
520 during midday due to both a large enhancement of VCPs, but also lower emissions from mobile
521 sources. Over the entire dataset, VCPs and mobile sources are estimated to represent 54% and 25%
522 of the total anthropogenic VOCs, respectively (Fig. 10B).

523

524 The cooking-dominated factor represents 10-30% of the total primary VOC mass resolved by
525 PMF, depending on the time of day. The relative fraction of the cooking-dominated factor peaks
526 in the mid-afternoon, as well as in the evening and night when activity along the Las Vegas Strip
527 is highest. Similar behavior has been observed in the relative abundance of primary cooking
528 organic aerosol in cities such as Los Angeles (Hayes et al., 2013). The cooking-dominated factor
529 is estimated to represent as much as 21% of the total anthropogenic VOCs over the entire dataset.

530

531 The fraction of cooking VOCs estimated here (21%) is specific to a site downwind of the Las
532 Vegas Strip where restaurants are abundant. These impacts are likely to vary across urban areas
533 based on ground site locations, restaurant density, and differences in the proportions of fossil fuel
534 and VCP emissions. Cooking emissions are commonly resolved from the source apportionment of
535 organic aerosol measurements in US cities (e.g., Hayes et al., 2013; Lyu et al., 2019; Zhang et al.,
536 2019; Xu et al., 2015). Far fewer studies have estimated the impact of cooking on the outdoor VOC
537 burden in US urban areas. Recently, cooking emissions were identified from source apportionment
538 of thermal desorption aerosol gas chromatograms and shown to be present at significant mixing
539 ratios in Livermore, CA (Wernis et al., 2022). Similarly, source apportionment of PTR-ToF-MS
540 data from Atlanta, GA shows that cooking emissions mixed with biomass burning were
541 responsible for 6–15% of the reported VOC carbon, which included contributions from fossil fuel,
542 VCPs, and biogenic sources (Peng et al., 2022). These proportions are similar to those reported
543 here, suggesting that cooking VOCs represent a significant fraction of total anthropogenic VOCs
544 in other US cities. Table S1 summarizes the cooking profile resolved by the PMF analysis. This
545 profile could be used to compare against measurements of cooking VOCs in other urban areas.



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Figure 10: (A) Diurnal contribution of VCP, mobile sources, and cooking factors to the sum of primary emissions apportioned by PMF (= VCP + mobile source + cooking). (B) Average contribution of the VCP, mobile source, and mobile source factors to the PMF solution. (C) Distribution of anthropogenic VOC emissions from the 2020 National Emissions Inventory for Clark County, NV. (D) Distribution of anthropogenic VOCs from FIVE-VCP-NEI17^{NRT} in Clark County, NV. The “other” category in both inventories reflect emissions from industry, farming, and electric power generation.

5. Comparison to Inventory Emissions

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The PMF results shown in Figure 10B are compared against the distribution of anthropogenic VOCs reported in emissions inventories used to model air quality (Figure 10, panels C and D). Panel C shows emissions reported in the 2020 National Emissions Inventory (NEI) for Clark County, NV. The NEI is a benchmark for determining US emissions standards and its methodology is fully described by the US Environmental Protection Agency (EPA, 2023). The NEI for Clark County includes emissions for mobile sources (e.g., on- and offroad vehicles), fossil fuel evaporative sources (e.g., gasoline stations), solvent evaporative sources (e.g., VCPs), and miscellaneous point and area sources. Cooking emissions in the NEI predominantly result from commercial sources with minor contributions from residential backyard barbecuing.



566

567 Panel D shows the distribution of VOCs represented by the FIVE-VCP-NEI17^{NRT} inventory
568 described by He et al. (2023). The FIVE-VCP inventory was developed following the methods
569 prescribed by McDonald et al. (2018) and was recently used to determine VCP impacts on air
570 quality in US cities (e.g., Coggon et al., 2021; Qin et al., 2021). He et al. (2023) updated FIVE-
571 VCP to include 2017 NEI emissions (NEI17) and revised VOC emissions with near real-time
572 (NRT) adjustment factors to account for COVID-19 impacts on various emission sectors. Mobile
573 source emissions are determined from fuel sales and on-road and off-road emission factors. VCP
574 emissions are estimated based on the mass balance of the chemical product industry for 2010 then
575 adjusted to 2021 emissions based on the long-term declining trends in VCP emissions reported by
576 Kim et al. (2022) and economic scaling factors reported by He et al. (2023). Other sectors are from
577 the NEI17 and are similarly updated with near real-time adjustment factors. Cooking emissions in
578 FIVE-VCP-NEI17^{NRT} are the same as those used in the 2017 NEI.

579

580 The 2020 NEI and FIVE-VCP-NEI17^{NRT} inventories both indicate that VCPs are the dominant
581 VOC emission sources in Clark County. Fossil fuel emissions are the next largest source, though
582 differences between the two inventories are evident. In the NEI, total fossil fuel emissions (= on-
583 and offroad emissions + evaporative emissions) are 15% lower than VCP emissions. In FIVE-
584 VCP-NEI17^{NRT}, fossil fuel emissions are 57% lower than VCPs. These differences are reflected
585 in previous comparisons between the NEI and FIVE-VCP (Coggon et al., 2021; McDonald et al.,
586 2018). The PMF solution shows that fossil fuels are ~54% lower than VCPs, which is most
587 consistent with FIVE-VCP-NEI17^{NRT}. Zhu et al. (2023) show that FIVE-VCP speciation agrees
588 well with VOCs primarily emitted from fossil fuel and VCPs reported during SUNVEX and
589 RECAP-CA. Aldehydes, ethanol, and monoterpenes were underestimated which may point to the
590 importance of missing emission sources, such as cooking.

591

592 The fraction of total cooking VOC emissions represented in both inventories is significantly
593 lower than the fraction resolved by PMF. Commercial sources dominate the cooking emissions in
594 the 2017 and 2020 NEI and are estimated based on food consumption estimates and emission
595 factors derived from laboratory studies (e.g., Schauer et al., 1999). The differences between the
596 PMF results and what is reported in the inventories may be partially explained by the spatial scale
597 of the datasets – the inventories represent county-level emissions estimates, while the observations
598 are specific to a site strongly influenced by the Las Vegas Strip. Nevertheless, the magnitude of
599 the disconnect highlights the need for further analysis. It is possible that the emission factors and/or
600 consumption of oils, meats, and other foods are different from what is reflected in laboratory
601 studies.

602

603 **6. Conclusions**

604



605 Mobile laboratory and ground site measurements were analyzed to determine the importance
606 of cooking emissions on urban VOC composition in Las Vegas, NV. PTR-ToF-MS data show that
607 cooking is a significant source of long-chain aldehydes to urban air. Measurements of octanal and
608 nonanal are found to be useful markers to evaluate cooking emissions due to their abundance in
609 restaurant plumes and local enhancements in areas with high restaurant density. A comparison of
610 the mass spectra downwind of restaurants to those obtained in regions with significant commercial
611 cooking show similar distributions in aldehydes and fatty acids known to be emitted from
612 laboratory cooking experiments.

613

614 Based on a PMF analysis, it is estimated that cooking emissions represent as much as 20% of
615 the anthropogenic VOCs emitted to the atmosphere in Las Vegas, NV. It is expected that the
616 relative importance of cooking emissions in other cities will vary based on regional restaurant
617 density and the magnitude of other anthropogenic emissions including VCPs and mobile sources.
618 More work is needed to quantify cooking in other urban areas. Measurements from this study in
619 Pasadena, CA (Fig S12) and those conducted previously in Livermore, CA and Atlanta, GA show
620 that long-chain aldehydes are ubiquitous in urban air (~100 – 200 ppt) and modulated by
621 commercial and residential cooking (Wernis et al., 2022; Peng et al., 2022). The source
622 apportionment profiles determined here may be compared against other urban environments to
623 evaluate cooking in other cities.

624

625 The VOCs emitted from cooking are reactive and may contribute to the formation of ozone,
626 secondary organic aerosol, and other pollutants such as peroxyacyl nitrates (Bowman et al., 2003).
627 A review of VOC emissions inventories show that total cooking emissions (i.e., residential +
628 commercial cooking) are likely underrepresented in air quality models. Spatial patterns of long-
629 chain aldehydes suggest that more work is needed to quantify the magnitude of emissions from
630 commercial cooking, which are also important sources of primary urban SOA (Robinson et al.,
631 2018; Shah et al., 2018). PMF results in Las Vegas suggest that cooking emissions may be as
632 important to urban VOCs as mobile sources in regions with significant restaurant activity.

633

634 **Data Availability**

635

636 Data for SUNVEx and RE-CAP are available at the NOAA CSL data repository
637 (<https://csl.noaa.gov/projects/sunvex/>).

638

639 **Author Contribution**

640

641 MMC, CES, XL, JBG, AL, JP, HJB, KA, and CW conducted measurements during SUNVEx and
642 RE-CAP. CH, QZ, RHS, JH, ML, KS, and BC developed inventories used to compare against
643 observations. MMC and CW wrote the paper with contributions from all authors.

644

645 **Competing Interests**

646



647 The authors also have no other competing interests to declare.

648

649 **Acknowledgements**

650

651 MMC, CES, QZ, and RHS acknowledge support from the U.S. Environmental Protection Agency
652 (EPA) STAR program (grant # 84001001). The views expressed in this document are solely those
653 of the authors and do not necessarily reflect those of the Agency. EPA does not endorse any
654 products or commercial services mentioned in this publication. CW, MMC, CES, LX, JBG, and
655 AL acknowledge measurement funding from Clark County, NV (contract number 20-022001) and
656 the California Air Resources Board (contract number 20RD002). This work was supported in part
657 by the NOAA Cooperative Agreements with CIRES, NA17OAR4320101 and
658 NA22OAR4320151. The authors thank Paul Wennberg, John Seinfeld, and Ben Schulze for their
659 coordination of the Caltech ground site during RECAP-CA.

660

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