



32 VOCs increases the oxygen to carbon ratios (O:C) in the oxidation products and can form  
33 highly oxygenated organic molecules (HOMs). These low volatility products are found to play  
34 a key role in the formation and growth of SOA (Ehn et al., 2014; Öström et al., 2017; Bianchi  
35 et al., 2019; Brean et al., 2019, 2020). However, heterogeneous and multiphase chemistry  
36 involving the reactions of organic compounds directly onto solid particles or inside liquid  
37 particles can also be an important contributor to SOA mass (Ervens et al., 2011, 2014; Kuang  
38 et al., 2020; Gu et al., 2023). The SOA is a dominant component of tropospheric fine particulate  
39 matter (Hallquist et al., 2009; Spracklen et al., 2011; Huang et al., 2014), influences oxidative  
40 capacity, local and global air quality, climate change, and human health (Jacobson et al., 2000;  
41 Hansen and Sato, 2001; Kanakidou et al., 2005; Zhang et al., 2014). Despite having significant  
42 attention due to its importance on SOA, its sources and formation processes are yet to be fully  
43 understood.

44 Aldehydes are common emissions in natural and polluted environments and have both  
45 biogenic and anthropogenic sources (Lipari et al., 1984; Carlier et al., 1986; Ciccioli et al.,  
46 1993; Schauer et al., 1999a, 1999b, 2001) and are also formed by chemical transformation of  
47 other VOCs, especially ozonolysis of alkenes (Calogirou et al., 1999). Atmospheric  
48 degradation of aldehydes is mainly governed by photolysis and the reaction with OH radicals  
49 in the daytime (Mellouki et al., 2003; Calvert et al., 2011; Mellouki et al., 2015). During  
50 nighttime, the reactions with NO<sub>3</sub> radicals are the dominant sink of aldehydes (Calvert et al.,  
51 2011). Although there are prior kinetic studies of reactions of *n*-aldehydes with OH radicals  
52 available in the literature (Albaladejo et al., 2002; Cassanelli et al., 2005; Iuga et al., 2010;  
53 Castañeda et al., 2012; S. Wang et al., 2015; Aguirre et al., 2025), they are limited to the initial  
54 steps of oxidation except recent studies (Barua et al., 2023; Yang et al., 2024) showing further  
55 oxidation steps leading to the formation of more functionalized products including HOMs.

56 It is well understood that the reaction of aldehydes with OH radicals is predominantly  
57 initiated by the abstraction of aldehydic hydrogen atom due to its weaker bond strength.  
58 However, with the increase of carbon chain length, the abstraction of other hydrogen atoms  
59 distant from the aldehydic moiety can also contribute to the overall oxidation process. The  
60 aldehydic H abstraction can lead to the cleavage of that carbon (C1) by CO loss from acyl  
61 (RC(O)) intermediate (Rissanen et al., 2014; Barua et al., 2023). Alternatively, it leads to the  
62 acyl peroxy (RC(O)OO) and, in high NO<sub>x</sub> (NO + NO<sub>2</sub>) condition, subsequently to acyloxy  
63 (RC(O)O) intermediate followed by CO<sub>2</sub> loss ultimately forming C<sub>*n*-1</sub> aldehyde, C<sub>*n*-1</sub> alkyl  
64 nitrate, and C<sub>*n*-1</sub> alkoxy isomerization products (Vereecken & Peeters, 2009; Chacon-Madrid  
65 et al., 2010). Besides, it can also produce C<sub>*n*</sub> peroxyacyl nitrates (PAN) via RC(O)OO + NO<sub>2</sub>

66 reaction (Mellouki et al., 2003; Chacon-Madrid et al., 2010; Calvert et al., 2011; Mellouki et  
67 al., 2015), a reservoir species for long-range transport of NO<sub>x</sub> in the free troposphere.  
68 Additionally, C<sub>n</sub> peroxy acids can be formed by the reaction of RC(O)OO intermediate and  
69 HO<sub>2</sub> (Barua et al., 2023).

70 Chacon-Madrid et al. (2010) have conducted a comparative study of the SOA yields in  
71 OH-initiated oxidation of *n*-aldehydes and *n*-alkanes under high NO<sub>x</sub> conditions. They reported  
72 near identical SOA yields from *n*-tridecanal and *n*-dodecane where the precursor alkane is  
73 having one less carbon than the precursor aldehyde. The finding was attributed to the formation  
74 of C<sub>12</sub> alkoxy radical intermediate from both precursors undergoing similar subsequent  
75 reactions leading to SOA. This indicates that the dominant fate of aldehyde oxidation by OH  
76 leads to the fragmentation of its carbon backbone losing one carbon atom rather than producing  
77 functionalized products with the same number of carbon atoms as the parent molecule.

78 In low NO<sub>x</sub> conditions, previous experimental studies have shown that the abstraction  
79 of the aldehydic hydrogen can also lead to molecular functionalization forming HOMs (Ehn et  
80 al., 2014; Rissanen et al., 2014; Tröstl et al., 2016; Z. Wang et al., 2021) and potentially  
81 promote the SOA yields. Recently, Barua et al. (2023) have studied hexanal OH oxidation  
82 reaction in detail using high level quantum chemical computations as well as experimental  
83 mass spectrometry technique. They showed that both the aldehydic and non-aldehydic H  
84 abstraction pathways can contribute to the functionalization of hexanal resulting in rapid  
85 formation of HOMs via autoxidation. Autoxidation refers to chain radical processes, generally  
86 starting with an oxygen-centered radical that undergoes unimolecular isomerization reaction  
87 leading to a carbon-centered radical species whose dominant fate is to add additional molecular  
88 oxygen and thus increases product O:C ratios (Crouse et al., 2013; Jokinen et al., 2014;  
89 Rissanen et al., 2014; Berndt et al., 2015; Mentel et al., 2015; Rissanen et al., 2015; Berndt et  
90 al., 2016). Along the aldehydic H abstraction reaction route, the fastest isomerization (1,6 H-  
91 shift rate coefficient,  $k = 0.2 \text{ s}^{-1}$ ) of the RC(O)OO intermediate was shown to be the key for  
92 autoxidation reaction chain propagation and competitive with any bimolecular reaction  
93 mediated RC(O)O fragmentation (Barua et al., 2023). Thus, the RC(O)OO isomerization  
94 reaction (Seal et al., 2023) keeps the carbon backbone of the precursor aldehyde intact. A non-  
95 aldehydic H abstraction from C4 was also seen to be competitive and its corresponding RO<sub>2</sub>  
96 was shown to undergo a 1,6 H-shift reaction with the aldehydic H atom at a higher rate ( $k =$   
97  $0.9 \text{ s}^{-1}$ ) than the H-shifts in RC(O)OO radical. The fast aldehydic H-shift to the peroxy group  
98 is consistent with other carbonyl systems reported previously (Da Silva, 2011; Crouse et al.,  
99 2012; Møller et al., 2016, 2019). Moreover, the kinetic modelling simulation on OH-initiated

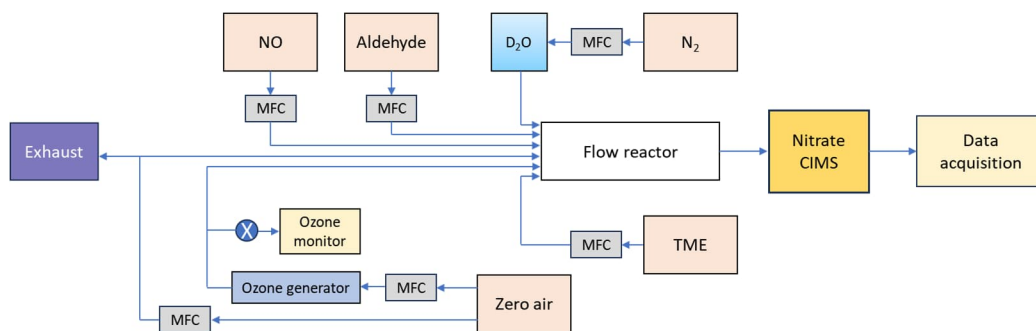
100 oxidation of hexanal conducted by Barua et al. (2023) showed that a detectable concentration  
101 ( $1.3 \times 10^4$  molecules  $\text{cm}^{-3}$ ) of  $\text{O}_7$  HOMs were produced even in the presence of 1 ppb NO.  
102 Because the non-aldehydic H abstraction pathways are likely less prone to fragmentation and  
103 promote functionalization, the effect of carbon chain length of *n*-aldehydes on HOM yields is  
104 of great interest.

105 In this work, we experimentally studied the OH initiated autoxidation of  $\text{C}_5$ – $\text{C}_8$  *n*-  
106 aldehydes in variable reaction times under atmospheric pressure and room temperature using  
107 state-of-the-art mass spectrometry technique. In addition, the reactions were studied in the  
108 presence of variable concentrations of NO to examine the effect of NO on the oxidation  
109 process. The study shows how the length of carbon chain in linear aldehydes directly affects  
110 the reactivity and functionalization of the molecules during their oxidation initiated by OH  
111 radicals.

## 112 ■ METHODS

### 113 2.1 Experimental setup

114 The gas-phase oxidation reactions of *n*-aldehydes with OH radicals were conducted in a flow  
115 reactor setup in the laboratory as shown in Fig. 1. All the experiments were conducted at room  
116 temperature and 1 atm pressure of air. The precursor aldehydes were introduced to the reactor  
117 from their individual gas cylinders. The oxidant OH radicals were produced in situ by the  
118 reaction of tetramethylethylene (TME,  $\text{C}_6\text{H}_{12}$ ) with ozone. We used an ozone generator that  
119 photolyzes zero-air by a mercury lamp (UVP, Analytik Jena) to provide ozone to the reactor  
120 while TME was supplied from a gas cylinder. The zero-air was produced by feeding  
121 compressed clean air to a zero-air generator (AADCO-737-15) which was also used as bath  
122 gas in the reactor maintaining a total sample flow of 8–10 slpm. The initial concentrations of  
123 reactant precursors were determined by controlling the individual gas flows using calibrated  
124 mass flow controllers (Alicat Scientific). An ozone analyzer (2B Technologies model 205) was  
125 used to measure the ozone concentrations. Further details regarding reactant concentration  
126 measurements, as well as specifications for the chemicals and gas cylinders, are provided in  
127 Sects. S1 and S2 of the Supplement.



128

129 **Figure 1.** Schematic representation of a flow reactor setup showing a nitrate ( $\text{NO}_3^-$ ) based  
 130 chemical ionization mass spectrometer coupled to ambient pressure flow reactor. TME =  
 131 tetramethylethylene ( $\text{C}_6\text{H}_{12}$ ). The oxidant OH radical was produced in situ by TME +  $\text{O}_3$   
 132 reaction. MFC = mass flow controller.

133 We studied the reactions over a range of reaction times (short: 1–3 s, and long: 11–13  
 134 s). The experimental conditions are presented in Table 1. Long reaction time experiments were  
 135 conducted using a borosilicate flow reactor (length: 100 cm, and i.d.: 4.7 cm) while a quartz  
 136 flow reactor (length: 100 cm, and i.d.: 2.2 cm) was used for the short reaction time experiments.  
 137 We utilized the full volume of the reactor to achieve a long reaction time. However, the short  
 138 reaction times were achieved by controlling the distance between the mass spectrometer orifice  
 139 and the position where the precursor aldehyde meets the oxidant OH inside the reactor. This  
 140 was done by using a movable injector that brings the aldehyde of interest at variable positions  
 141 inside the reactor. The shortest possible reaction time of an individual aldehyde was chosen by  
 142 the detection of any HOMs from its oxidation initiated by OH radicals. In short reaction time  
 143 experiments, the highest concentration of VOC (6.4 ppmv) was used for pentanal while the  
 144 concentrations of other aldehydes were up to 1 ppmv. The VOC concentrations of 0.2–2.5  
 145 ppmv were used in long reaction time experiments. The other reactants including 43–97 ppbv  
 146 of TME, and 77–295 ppbv of ozone were maintained nearly constant with respect to individual  
 147 VOC in all experiment types (see Table 1). Among all the studied *n*-aldehyde systems, the  
 148 lowest level of aldehyde and  $\text{O}_3$  concentrations were maintained for heptanal oxidation  
 149 experiment. This is because higher concentrations led to irrelevant products likely originating  
 150 from the ozonolysis of heptanal stabilizers (see Fig. S2 in the Supplement). To observe the  
 151 effect of  $\text{NO}_x$  on the OH induced oxidation of *n*-aldehydes, we conducted the experiments with  
 152 variable  $\text{NO}$  concentrations (2–1000 ppbv). Additionally, one more set of experiments,  
 153 hydrogen to deuterium (H/D) exchange, were conducted by the addition of  $\text{D}_2\text{O}$  flow to the *n*-  
 154 aldehyde OH oxidation reaction. These experiments give an estimate of the number of  
 155 functional groups with labile H atoms (e.g., OH, OOH, and  $\text{C}(\text{O})\text{OOH}$ ) in the oxidation

156 products. A near complete H/D conversion was confirmed by monitoring the reagent ion  
157 signals of  $\text{HNO}_3\text{NO}_3^-$  and  $(\text{HNO}_3)_2\text{NO}_3^-$  fully converting to  $\text{DNO}_3\text{NO}_3^-$  and  $(\text{DNO}_3)_2\text{NO}_3^-$ ,  
158 respectively (see Fig. S14 in the Supplement). The time series of reactive species, VOC, OH,  
159  $\text{RO}_2$ ,  $\text{O}_3$ , and NO under different reaction conditions are shown in Figs. S16–S17 in the  
160 Supplement.

161 The oxidation products were detected using a nitrate ion time-of-flight chemical  
162 ionization mass spectrometer ( $\text{NO}_3^-$ -ToF-CIMS) as their  $\text{NO}_3^-$  adducts. A zero-air sheath flow  
163 of 20 slpm was provided to the chemical ionization inlet. The  $\text{NO}_3^-$  ions were produced from  
164 gas-phase  $\text{HNO}_3$  flow under soft X-ray exposure while being carried by  $\text{N}_2$  to the inlet. The  
165 mass spectrometric data processing, including averaging, baseline removal, mass axis  
166 calibration, and peak integration were done using the tofTools v6.03 package for MATLAB.

## 167 **2.2 Kinetic simulation**

168 We estimated the concentrations of reactive species in the flow reactor during different *n*-  
169 aldehyde oxidation experiments using a kinetic simulator Kinetiscope (version 1.1.1136.x64)  
170 (Hinsberg and Houle, 2022). These include the average concentrations of OH radicals and  
171 initial  $\text{RO}_2$  radicals (i.e.,  $\text{C}_n\text{H}_{2n-1}\text{O}_3$ , the first peroxy radicals formed in the oxidation process)  
172 produced in the experiments. In the simulations, a single-reactor model with constant volume,  
173 pressure, and temperature was employed. The temperature was set to 298.15 K. The simulation  
174 setting parameters such as total number of particles ( $1 \times 10^9$ ) and random number seed  
175 (12947) were kept constant for consistency, while the maximum simulation time was matched  
176 to that of individual experiments. Details of all the simulations are provided in Sect. S13 in the  
177 Supplement.

178 **Table 1.** The experimental conditions for OH initiated oxidation of studied C<sub>5</sub>–C<sub>8</sub> *n*-aldehydes.

Experiment type	[VOC]	[TME]	[O <sub>3</sub> ]	[OH] <sup>‡</sup>	[NO]	D <sub>2</sub> O	$\Delta t$ <sup>‡</sup>
VOC	ppmv	ppbv	ppbv	pptv	ppbv	y/n <sup>±</sup>	s
<b>Short residence time</b>							
Pentanal (C <sub>5</sub> H <sub>10</sub> O)	6.4	48.2	295	4.4	N/A <sup>†</sup>	n	2.3
Hexanal (C <sub>6</sub> H <sub>12</sub> O)	1.0	43.2	225	3.4	N/A	n	1.1, 2.9
Octanal (C <sub>8</sub> H <sub>16</sub> O)	0.72	48.2	208	3.1	N/A	n	1.0, 2.1
<b>Long residence time</b>							
Pentanal (C <sub>5</sub> H <sub>10</sub> O)	2.5	48.2	295	4.4	N/A	n	12.8
Hexanal (C <sub>6</sub> H <sub>12</sub> O)	1.0	43.2	225	3.4	N/A	n	11.5
Heptanal (C <sub>7</sub> H <sub>14</sub> O)	0.15	96.5	77	1.2	N/A	n	12.8
Octanal (C <sub>8</sub> H <sub>16</sub> O)	0.72	48.2	208	3.1	N/A	n	12.8
<b>Experiments with NO</b>							
Pentanal (C <sub>5</sub> H <sub>10</sub> O)	1.3	48.2	208	3.1	2–1000	n	12.8
Hexanal (C <sub>6</sub> H <sub>12</sub> O)	1.0	43.2	225	3.4	2–200	n	11.5
Octanal (C <sub>8</sub> H <sub>16</sub> O)	0.72	48.2	208	3.1	2–1000	n	12.8
<b>Experiments with D<sub>2</sub>O</b>							
Pentanal (C <sub>5</sub> H <sub>10</sub> O)	2.5	48.2	295	4.4	N/A	y	12.8
Hexanal (C <sub>6</sub> H <sub>12</sub> O)	1.0	43.2	225	3.4	N/A	y	11.5
Heptanal (C <sub>7</sub> H <sub>14</sub> O)	0.10	96.5	77	1.2	N/A	y	12.8
Octanal (C <sub>8</sub> H <sub>16</sub> O)	0.72	48.2	208	3.1	N/A	y	12.8

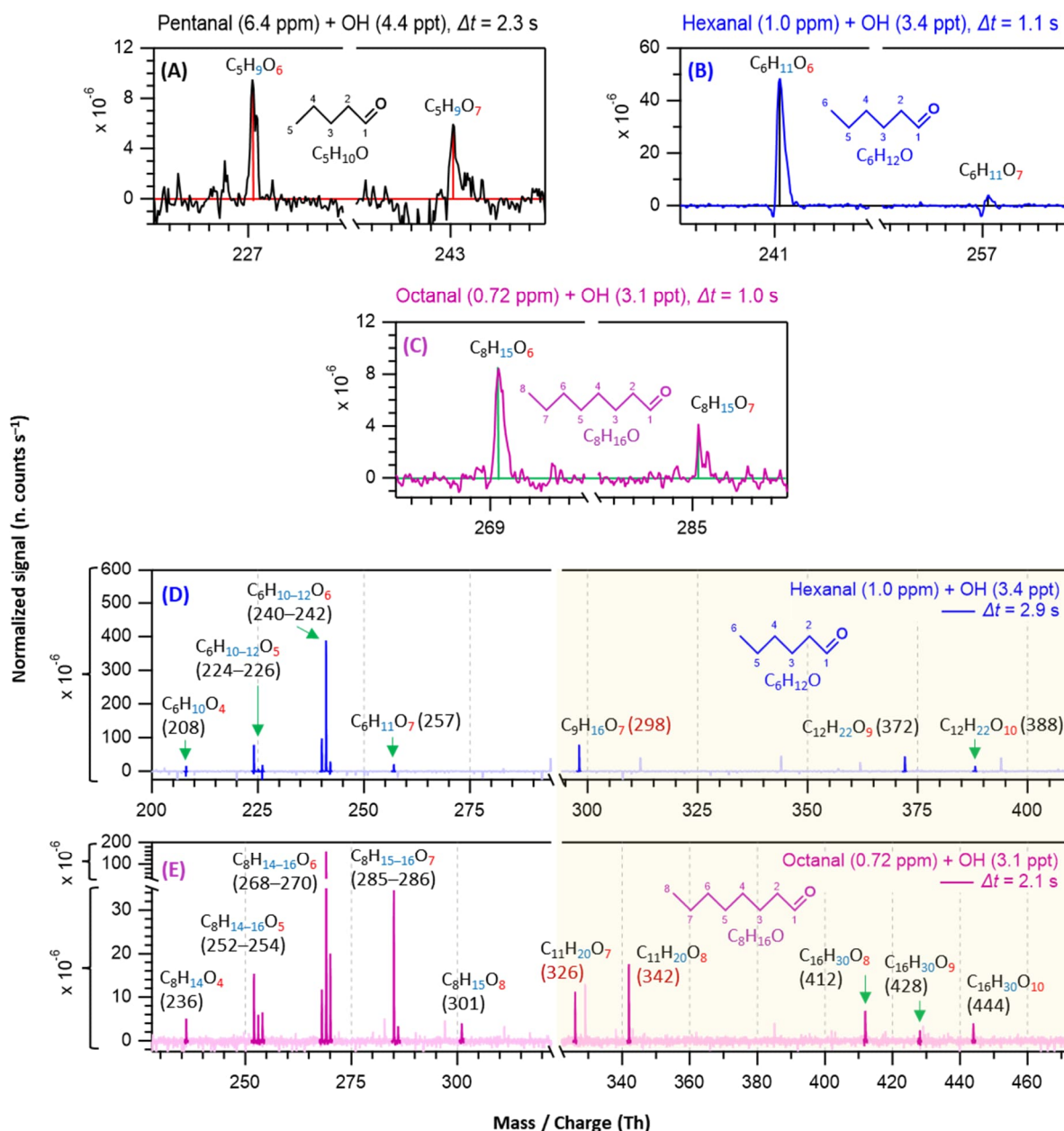
179 <sup>‡</sup> The initial OH concentrations were calculated using bimolecular rate coefficients  $k_{O_3-TME} =$   
180  $1.5 \times 10^{-15} \text{ cm}^3 \text{ s}^{-1}$ ,  $k_{OH-TME} = 1.0 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$  (Manion et al., 2015), and the expression  
181  $[OH] = (k_{O_3-TME} * [O_3]) / k_{OH-TME}$ .

182 <sup>†</sup> Not applicable (N/A). <sup>±</sup> D<sub>2</sub>O added = y, not added = n. <sup>‡</sup> Reaction time ( $\Delta t$ ).

## 183 ■ RESULTS AND DISCUSSION

### 184 3.1 Detection of HOM in short to long reaction time experiments

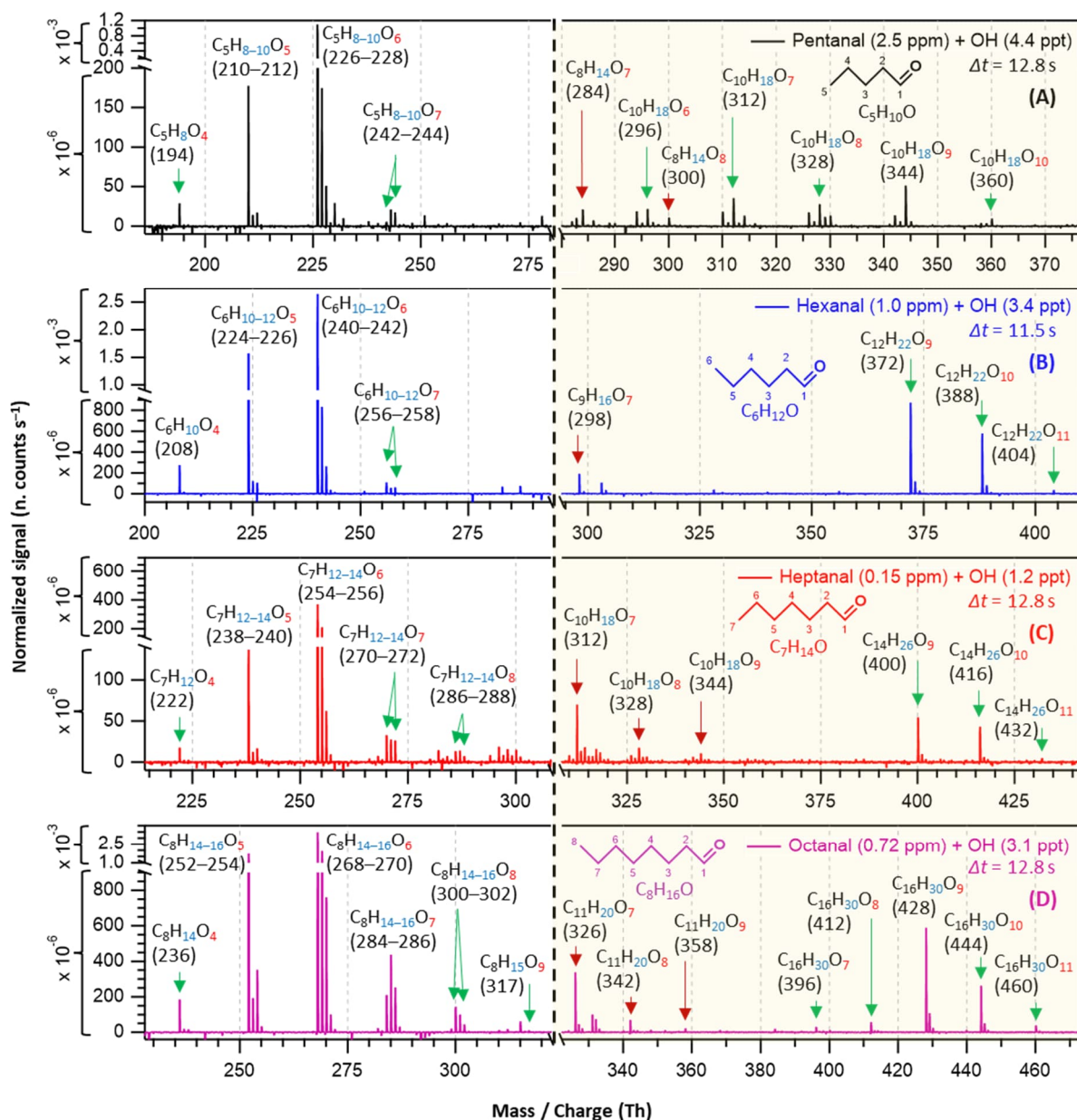
185 In this section, we discuss how early HOMs formed and how they evolved with the progress  
186 of reaction time in different *n*-aldehyde OH oxidation experiments. The hexanal OH oxidation  
187 spectra included in Figs. 2–3 are reproduced from our previous study (Barua et al., 2023) for  
188 comparison with the other aldehydes. Figure 2 shows the results from short reaction time ( $\Delta t =$   
189 1–3 s) experiments. We observed the formation of O<sub>6</sub> and O<sub>7</sub> HOMs within 1.0, 1.1, and 2.3 s  
190 reaction times in the oxidation experiments of octanal, hexanal, and pentanal, respectively. It  
191 is essential to mention that with the increase of number of carbon atoms in the studied  
192 aldehydes, the required precursor concentrations for first HOM observation decreased (from  
193 6.4 ppm pentanal, 1 ppm hexanal to 0.72 ppm octanal; corresponding reacted concentrations  
194 1.15, 0.34, and 0.30 ppb, respectively). This shows a clear effect of carbon chain length on the  
195 reactivity of linear aldehydes towards HOM formation. Because the oxidation process of *n*-  
196 aldehydes (C<sub>*n*</sub>H<sub>2*n*</sub>O) with OH is initiated by the abstraction of a H atom (aldehydic or non-  
197 aldehydic), the first formed acyl (or alkyl) peroxy radical C<sub>*n*</sub>H<sub>2*n*-1</sub>O<sub>3</sub> contains an odd number  
198 of oxygen atoms. If autoxidation outcompetes any other bimolecular reactions (e.g., RO<sub>2</sub> +  
199 RO<sub>2</sub>, RO<sub>2</sub> + HO<sub>2</sub>, etc.), the product spectrum will be mostly dominated by odd number of  
200 oxygen containing products. In all studied *n*-aldehyde systems, the intensity of O<sub>6</sub> HOM is  
201 higher than that of O<sub>7</sub> HOM (Figs. 2–3). The formation of C<sub>*n*</sub>H<sub>2*n*-1</sub>O<sub>6</sub> peroxy radical indicates  
202 that the process certainly involves a bimolecular reaction step. In hexanal OH oxidation, Barua  
203 et al. (2023) computationally showed that the formation of C<sub>6</sub>H<sub>11</sub>O<sub>5</sub> peroxy radical via  
204 autoxidation is very fast while the subsequent isomerization reaction leading to C<sub>6</sub>H<sub>11</sub>O<sub>7</sub> is  
205 slower. The same is likely to hold true for other aldehydes and it is expected that the C<sub>*n*</sub>H<sub>2*n*-1</sub>O<sub>5</sub>  
206 peroxy radical undergoes a bimolecular reaction converting it to C<sub>*n*</sub>H<sub>2*n*-1</sub>O<sub>4</sub> alkoxy radical,  
207 followed by a H-shift, and subsequent O<sub>2</sub> addition reactions producing the dominant C<sub>*n*</sub>H<sub>2*n*-1</sub>O<sub>6</sub>  
208 HOM (see Figs. S7–S11 in the Supplement). As the reaction time increased, we observed the  
209 formation of monomeric HOM up to O<sub>7</sub> and accretion products up to O<sub>10</sub> composition within  
210 2.9 s in hexanal oxidation (see Fig. 2D) with the consumption of  $\sim 8.88 \times 10^{-4}$  of its initial  
211 concentration. In the case of octanal, monomeric O<sub>8</sub> HOM and accretion products up to O<sub>10</sub>  
212 formed within 2.1 s reaction time (see Fig. 2E) with a comparable reacted fraction  
213 ( $\sim 8.71 \times 10^{-4}$ ) of its initial concentration.



214

215 **Figure 2.** Nitrate chemical ionization mass spectra of OH initiated oxidation of *n*-aldehydes  
 216 showing the formation of HOMs in different reaction times ( $\Delta t$ ): 2.3 s – pentanal in black (A),  
 217 1.1 and 2.9 s – hexanal in blue (B and D), and 1.0 and 2.1 s – octanal in purple (C and E). The  
 218 product peaks are labelled with the exclusion of  $\text{NO}_3^-$  ion attachment in their compositions.  
 219 The backgrounds of TME ozonolysis (TME +  $\text{O}_3$ ) and aldehyde have been subtracted from all  
 220 spectra, resulting in several negative peaks in panels A–E. The accretion product region is  
 221 highlighted in light gold background. The accretion products labeled with nominal mass/charge  
 222 in dark red ( $\text{C}_{n+3}\text{H}_{2n+4}\text{O}_{7-8}$ ) are related to the TME-derived peroxy radical  $\text{C}_3\text{H}_5\text{O}_3$ .

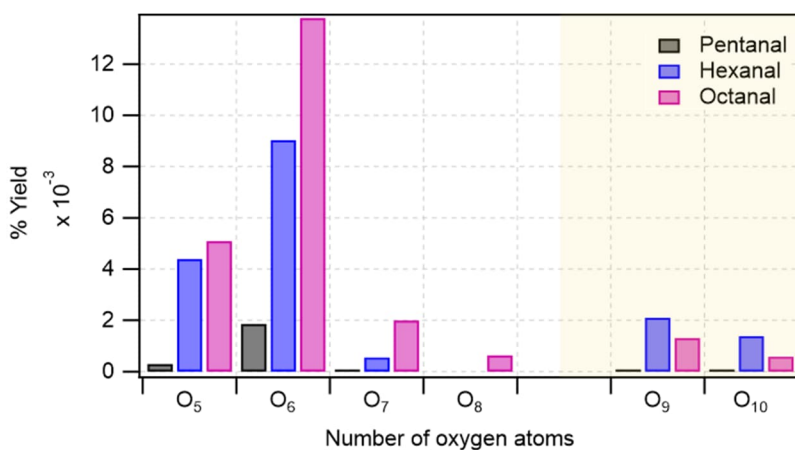
223 In long reaction time (11–13 s) experiments, we observed higher intensities of product  
224 signals (see Fig. 3) in comparison to their intensities in the short reaction time experiments - as  
225 expected. For pentanal, Fig. 3A shows that HOM accretion products up to O<sub>10</sub> formed within  
226 12.8 s reaction time which were not seen in the short reaction time experiment (2.3 s). A lower  
227 precursor concentration, 2.5 ppm of pentanal in long reaction time experiment compared to  
228 earlier 6.4 ppm in short reaction time, was sufficient to produce the observed HOMs in this  
229 case. A close observation of C<sub>6</sub>–C<sub>8</sub> *n*-aldehyde oxidation spectra (Fig. 3B–D) reveals that HOM  
230 accretion products up to O<sub>11</sub> formed within 11–13 s reaction time under the experimental  
231 conditions. In all *n*-aldehyde experiments, we also observed the accretion products resulting  
232 from different combinations of aldehyde-derived peroxy radicals C<sub>n</sub>H<sub>2n-1</sub>O<sub>6-8</sub> and TME-  
233 derived peroxy radical C<sub>3</sub>H<sub>5</sub>O<sub>3</sub> (see Fig. S13 in the Supplement for details) which are marked  
234 with dark red arrows. Figure 3D implies that the highest oxygenation (C<sub>8</sub>H<sub>15</sub>O<sub>9</sub>) in the  
235 monomeric HOM products is associated with octanal, whereas Fig. 3C shows that the most  
236 oxygenated products produced from heptanal are C<sub>7</sub>H<sub>12-14</sub>O<sub>8</sub>. In the case of pentanal and  
237 hexanal, monomeric HOMs are limited to seven oxygen atoms (see Fig. 3A–B). All in all, we  
238 notice a near identical distribution of oxidation products in the experiments with all C<sub>5</sub>–C<sub>8</sub> *n*-  
239 aldehydes. However, the tendency of oxidation gets faster and advances to higher oxygenated  
240 products when the carbon chain length increases.



241

242 **Figure 3.** Nitrate chemical ionization mass spectra of OH initiated oxidation of *n*-aldehydes in  
 243 11–13 s residence times: pentanal in black (A), hexanal in blue (B), heptanal in red (C), and  
 244 octanal in purple (D). The product peaks are labelled with the exclusion of  $NO_3^-$  ion attachment  
 245 in their compositions. The backgrounds of TME ozonolysis (TME +  $O_3$ ) and aldehyde have  
 246 been subtracted from all spectra, resulting in several negative peaks in panels A–D. The  
 247 accretion product region is highlighted in light gold background. The accretion products  
 248 marked by dark red arrows ( $C_{n+3}H_{2n+4}O_{7-9}$ ) are related to the TME-derived peroxy radical  
 249  $C_3H_5O_3$ .

250 The bar plot (see Fig. 4) compares the yields of major oxidation products from different  
 251 *n*-aldehydes. It clearly shows that the yields of higher oxygenated products increase as we move  
 252 from pentanal to octanal. In the accretion product regime, the yields of O<sub>9</sub>–O<sub>10</sub> products in  
 253 octanal are lower than that of hexanal which is also reflected in their dimer to monomer ratios  
 254 with octanal being  $8.8 \times 10^{-2}$  and hexanal being  $2.5 \times 10^{-1}$ . The lower ratio for octanal  
 255 compared to hexanal is observed despite both precursors producing comparable quantities of  
 256 initial RO<sub>2</sub> radicals ( $9.12 \times 10^9$  and  $9.37 \times 10^9$  molecules cm<sup>-3</sup> from octanal and hexanal,  
 257 respectively; see Table S1 in the Supplement). This can lie in the variation of RO<sub>2</sub> + RO<sub>2</sub>  
 258 reaction rate coefficients forming the accretion products (RO<sub>2</sub> + RO<sub>2</sub> → ROOR + O<sub>2</sub>) which is  
 259 highly dependent on specific RO<sub>2</sub> structures (Shallcross et al., 2005; Berndt et al., 2018).

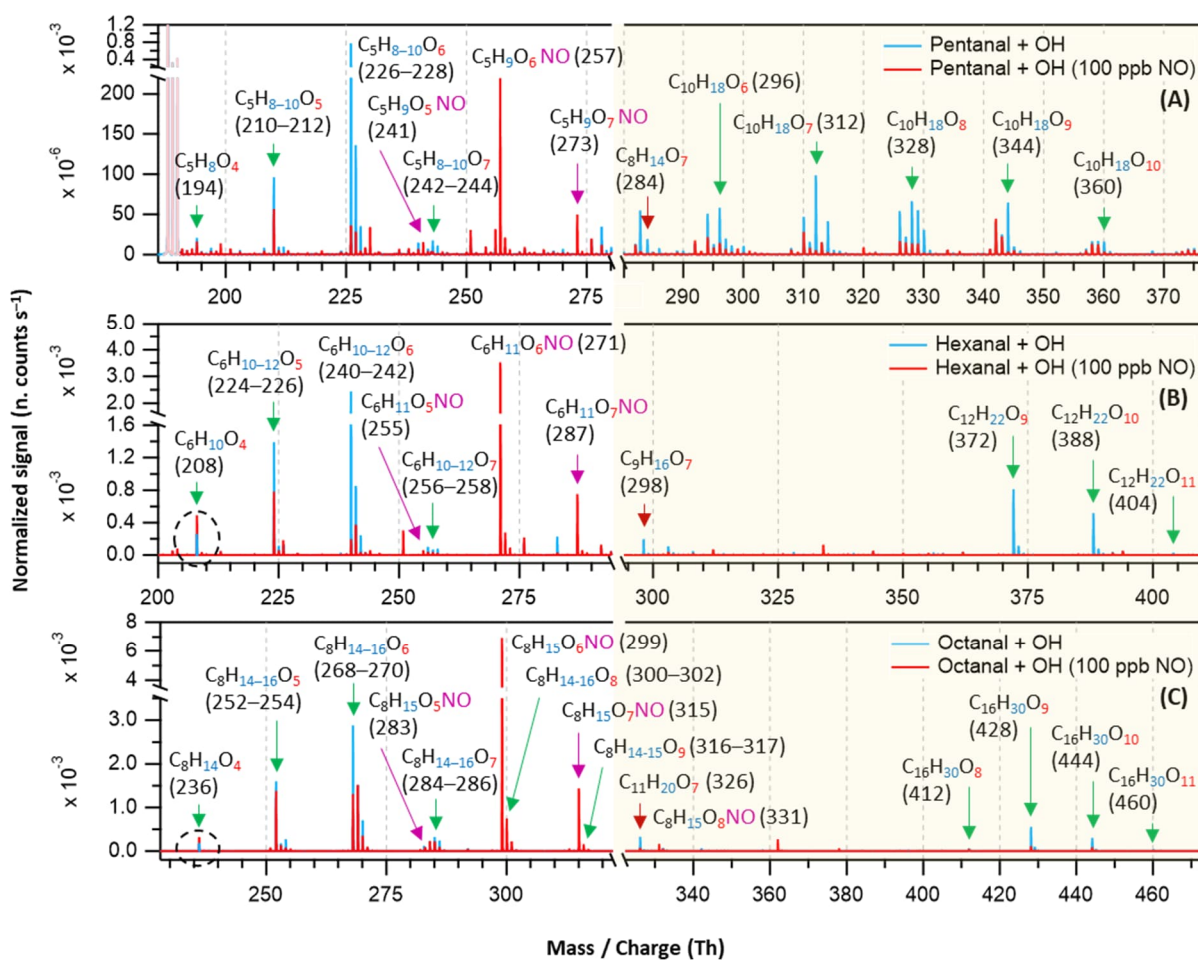


260  
 261 **Figure 4.** Distribution of major oxidation products (O<sub>5</sub>–O<sub>8</sub> in monomeric regime with white  
 262 background, and O<sub>9</sub>–O<sub>10</sub> in accretion product regime with light gold background) in pentanal,  
 263 hexanal, and octanal oxidation initiated by OH radical. In y-axis, the numbers are the  
 264 cumulative sum of yields of products with the same oxygen number. Reaction time,  $\Delta t = 11$ –  
 265 13 s.

### 266 3.2 Experiments in the presence of NO

267 It has been widely acknowledged that the formation of HOM is suppressed in high NO<sub>x</sub>  
 268 conditions (Wildt et al., 2014; Praske et al., 2018; McFiggans et al., 2019; Pullinen et al., 2020),  
 269 thus reducing the SOA yields. In this process, the reduction in SOA yield is largely attributed  
 270 to the suppression of highly condensable HOM accretion products (RO<sub>2</sub> + RO<sub>2</sub> → ROOR)  
 271 (Kirkby et al., 2016; Pullinen et al., 2020). However, other studies have shown that NO can  
 272 also enhance HOM formation by producing reactive RO radicals (RO<sub>2</sub> + NO → RO + NO<sub>2</sub>)  
 273 that can propagate autoxidation (Rissanen, 2018; Yan et al., 2020; Z. Wang et al., 2021; Shen  
 274 et al., 2022; Nie et al., 2023; Barua et al., 2025; Kang et al., 2025). In our different *n*-aldehyde

275 oxidation experiments in the presence of variable concentrations of NO, we observed the  
 276 general tendency of dropping of the accretion product signals ( $C_{2n}H_{4n-2}O_z$ ) and their  
 277 corresponding yields as expected (see Figs. 5 and 6). Figure 5 represents the mass spectra  
 278 recorded in the experiments without (in blue) and with the presence of 100 ppb initial NO (in  
 279 red). At this condition, most of the monomeric products are quenched at different extents  
 280 alongside the formation of organonitrates while the accretion products are quenched nearly  
 281 completely. Interestingly, we observed some enhancement in the intensities of  $O_4$  products in  
 282 the case of hexanal and octanal (Fig. 5B–C) under 100 ppb NO. A closer look at Fig. 6A reveals  
 283 that the yields of closed-shell products  $C_5H_8O_{4-6}$  somewhat increased under around 1 ppb  
 284 average NO condition which then started to decrease under higher NO of around 30 ppb and  
 285 above in pentanal oxidation. In both pentanal and hexanal oxidations, the dominant  $O_6$  peroxy  
 286 radicals ( $C_5H_9O_6$  and  $C_6H_{11}O_6$  respectively, blue markers) gained higher yields under around  
 287 30 ppb of NO (Fig. 6A–B) compared to without NO condition.

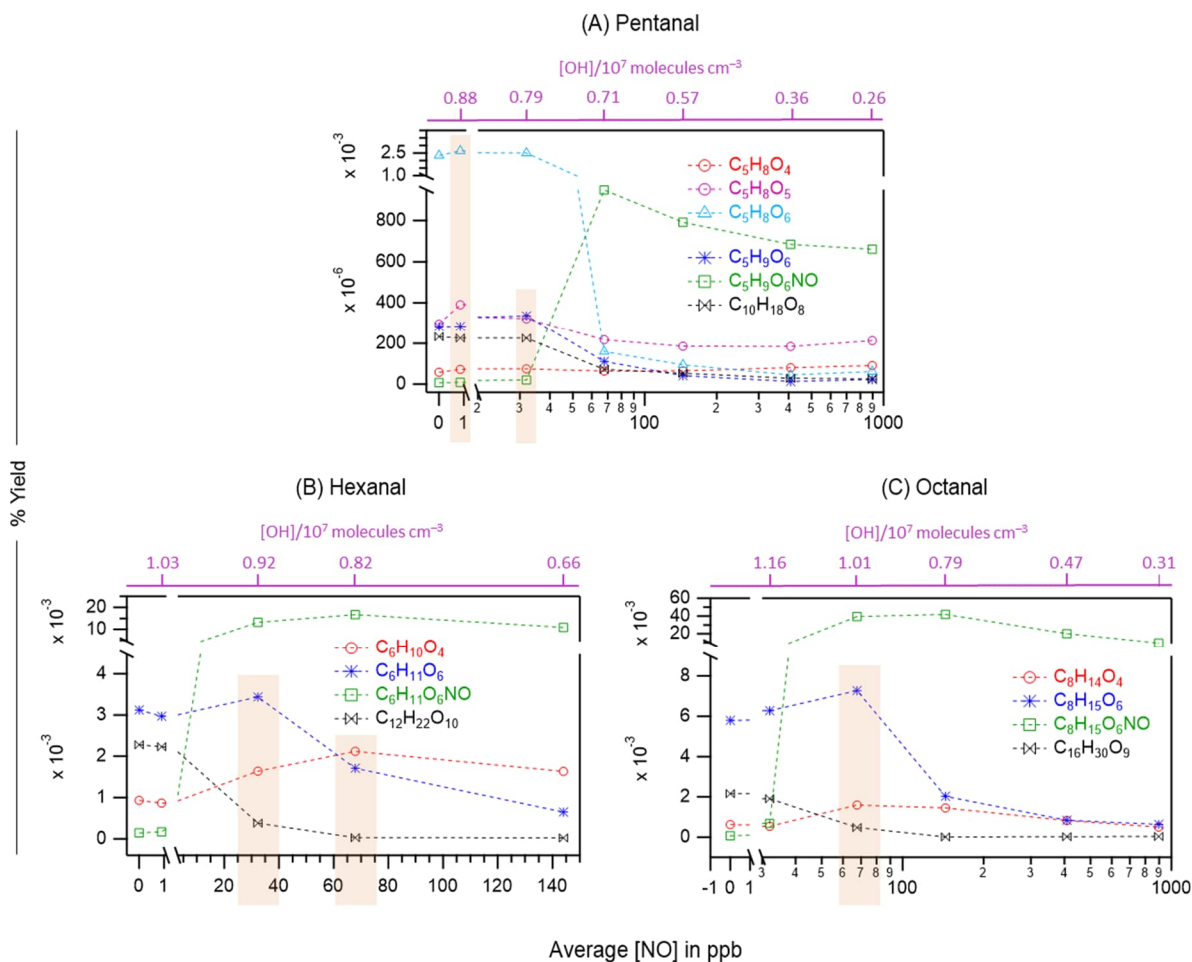


288

289 **Figure 5.** Overlaid nitrate chemical ionization mass spectra of OH initiated oxidation of *n*-  
 290 aldehydes without (in blue) and with the presence of 100 ppb of initial NO (in red): pentanal

291 (A), hexanal (B), and octanal (C). The product peaks are labelled with the exclusion of  $\text{NO}_3^-$   
 292 ion attachment in their compositions. The accretion product region is highlighted in light gold  
 293 background. The accretion products marked by dark red arrows ( $\text{C}_{n+3}\text{H}_{2n+4}\text{O}_{7-9}$ ) are related to  
 294 the TME-derived peroxy radical  $\text{C}_3\text{H}_5\text{O}_3$ . The organonitrates (in the presence of NO) are  
 295 labelled with the extension NO and marked with purple arrows. Reaction time,  $\Delta t = 11\text{--}13$  s.

296 On the other hand, in octanal oxidation, the yield of dominant  $\text{O}_6$  peroxy radical ( $\text{C}_8\text{H}_{15}\text{O}_6$ )  
 297 increased even under 70 ppb of NO (see Fig. 6C). These observations indicate that the  
 298 suppressing effect of NO on the yields of HOMs in *n*-aldehyde oxidation is perturbed with the  
 299 increase of carbon chain length of the precursor aldehyde. Also, in all the studied *n*-aldehydes,  
 300 although highly oxygenated products are suppressed under higher NO concentrations, we see  
 301 some enhancement in the early oxygenated closed shell products ( $\text{O}_4\text{--}\text{O}_5$ , and even  $\text{O}_6$  product  
 302 in pentanal) under relatively lower NO concentrations. It should be noted that the formation of  
 303 organonitrates with chemical composition  $\text{C}_n\text{H}_{2n-1}\text{O}_z\text{NO}$  strongly supports our assignment of  
 304 reactive peroxy radical intermediates  $\text{C}_n\text{H}_{2n-1}\text{O}_z$ . Additional insights into the abundance of  
 305 HOMs under varying NO concentrations are discussed in Sect. S12 in the Supplement.

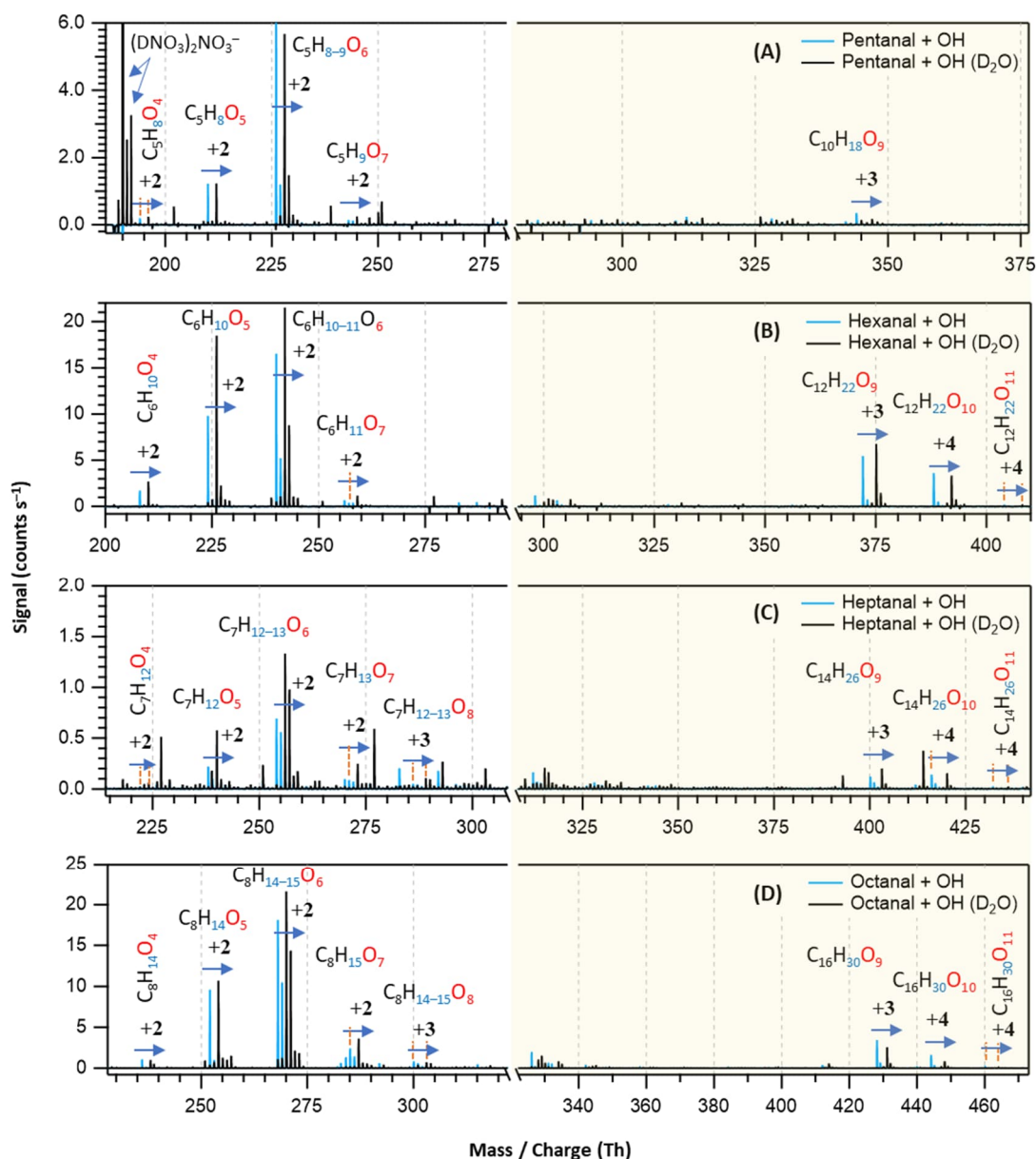


306

307 **Figure 6.** The yields of different oxidation products including monomeric HOMs,  
308 organonitrates (green markers), and HOM accretion products (black markers) as a function of  
309 average NO concentrations in OH initiated oxidation of *n*-aldehydes: pentanal (A), hexanal  
310 (B), and octanal (C). The corresponding average OH radical concentrations in these  
311 experiments are shown with purple scales. Note the logarithmic scale (x-axis) in panels A and  
312 C. The orange rectangles highlight the enhanced yields of several non-nitrogen containing  
313 products: O<sub>4</sub>–O<sub>6</sub> closed-shell products from pentanal under around 1 ppb NO (A), O<sub>4</sub> closed-  
314 shell products from hexanal and octanal under around 70 ppb NO (B and C), O<sub>6</sub> peroxy radical  
315 from pentanal and hexanal under around 30 ppb NO (A and B) and the same from octanal under  
316 70 ppb NO (C). Reaction time,  $\Delta t = 11\text{--}13$  s.

### 317 **3.3 D<sub>2</sub>O experiments**

318 With the addition of D<sub>2</sub>O in the OH initiated oxidation experiments of *n*-aldehydes, we  
319 observed a shift in individual product signals in the mass spectra equivalent to the number of  
320 exchangeable H atoms in the product structures (see Fig. 7). This provides additional insight  
321 into the product identities in terms of the total number of OH, OOH, and (or) C(O)OOH groups  
322 present in their molecular structures. Figure 7 shows that the oxidation products with same  
323 number of oxygen atoms in all the studied *n*-aldehydes undergo identical mass shifts (H/D  
324 exchange) in the presence of D<sub>2</sub>O. This observation indicates that the autoxidation mechanism  
325 derived by Barua et al. (2023) for hexanal OH oxidation (see Fig. S5 in the Supplement) is  
326 directly applicable to the other linear aldehydes studied here and thus produces similar product  
327 structures. The original mechanism is extended to HOMs up to nine oxygen atoms and  
328 presented in Fig. S6 in the Supplement. The likely formation process of HOM accretion  
329 products (C<sub>2n</sub>H<sub>4n-2</sub>O<sub>9-11</sub>) is shown in Fig. S12 in the Supplement. The proposed structures of  
330 O<sub>5</sub> closed-shell products, O<sub>6</sub>–O<sub>8</sub> monomeric HOMs as well as O<sub>9-11</sub> HOM accretion products  
331 (see Figs. S6–S12 in the Supplement) agree with the H/D exchange experiments in terms of  
332 their 2–4 units of mass shifts in the respective *n*-aldehyde mass spectra (see Fig. 7).



333  
 334 **Figure 7.** Overlaid nitrate chemical ionization mass spectra of OH initiated oxidation of *n*-  
 335 aldehydes without (in blue) and with the presence of D<sub>2</sub>O (in black): pentanal (A), hexanal (B),  
 336 heptanal (C), and octanal (D). In panel (A), the label (DNO<sub>3</sub>)<sub>2</sub>NO<sub>3</sub><sup>-</sup> is assigned to the deuterated  
 337 nitric acid trimer reagent signal. The product peaks are labelled with the exclusion of NO<sub>3</sub><sup>-</sup> ion  
 338 attachment in their compositions and the numbers on the blue arrows indicate the individual  
 339 counts of mass shift during H/D exchange. The backgrounds of TME ozonolysis (TME + O<sub>3</sub>)  
 340 and aldehyde have been subtracted from the spectra, resulting in several negative peaks in  
 341 panels A–B. The accretion product region is highlighted in light gold background.

### 342 **3.4 Atmospheric implications**

343 Ambient concentration of individual longer chain aldehyde ( $\geq C_5$ ) can vary from sub-ppb to  
344 several ppb depending on time and location (Williams et al., 1996; Duan et al., 2008; Li et al.,  
345 2018; Ma et al., 2019). A total concentration of  $C_6$ – $C_{10}$  *n*-aldehydes in Monti Cimini Forest in  
346 Italy was measured to be 8.8 ppb (Ciccioli et al., 1993). In indoor air, the concentration can be  
347 significantly higher, even around 50 ppb (Birmili et al., 2022). Atmospheric lifetime of *n*-  
348 aldehydes due to their reactivity with OH radicals is generally less than 10 h (Albaladejo et al.,  
349 2002; Aguirre et al., 2025). Because of their significant photochemical ozone formation  
350 potential (Jenkin et al., 2017; Aguirre et al., 2025), they are good candidates for generating  
351 photochemical smog in  $NO_x$  rich polluted urban atmosphere. On the other hand, previous  
352 studies have shown that atmospheric oxidation products of longer chain *n*-aldehydes are direct  
353 contributors to the formation of SOA (Chacon-Madrid et al., 2010; Fan et al., 2024). Here, we  
354 demonstrated that the studied  $C_5$ – $C_8$  *n*-aldehydes can rapidly form HOM via autoxidation  
355 initiated by OH radicals, and the length of carbon chain controls the efficiency of the process.  
356 Therefore, with the increase of carbon chain length in *n*-aldehydes, the fast formation of HOMs  
357 is expected to take part in the early stages of gas-to-particle formation and growth contributing  
358 to atmospheric SOA. Our experiments in the presence of NO showed a general decreasing trend  
359 of HOM accretion products with increasing NO but also formed the corresponding highly  
360 oxygenated organic nitrates (HOM-ONs). In our study with pentanal at 2 ppb NO condition,  
361 some enhancement with oxidation products up to six O atoms was also seen. Pullinen et al.  
362 (2020) showed that both HOMs and HOM-ONs originated from the same peroxy radicals with  
363 more than six O atoms condensed on particles by about 50% and those with more than eight O  
364 atoms condensed by about 100% to form SOA in monoterpene photooxidation. Moreover,  
365 other reports show that HOM-ONs originated from different VOCs can contribute to low  
366 volatility products (Barua et al., 2025) and thereby particle growth and aerosol mass loading  
367 (Fry et al., 2014; Lee et al., 2016; W. Huang et al., 2019).

### 368 **■ CONCLUSIONS**

369 This study represents the significance of longer chain linear aldehydes ( $\geq C_5$ ), key components  
370 of atmospherically abundant oxygenated volatile organic compounds (OVOCs), in rapid  
371 formation of HOMs upon atmospheric oxidation and their potential contribution to atmospheric  
372 SOA. Among the studied  $C_5$ – $C_8$  *n*-aldehydes, the fastest HOM formation is associated with  
373 octanal forming  $O_6$  and  $O_7$  HOMs within 1.0 s reaction time with low precursor loading.

374 Pentanal and hexanal formed HOMs with the same number of oxygen atoms as early as 2.3  
375 and 1.1 s, respectively but with higher precursor loadings (i.e., 6.4 ppm pentanal and 1.0 ppm  
376 hexanal compared to 0.72 ppm octanal). The highest oxygenated monomeric HOM with 9  
377 oxygen atoms was formed from octanal whereas the numbers are up to 8 oxygen atoms for  
378 heptanal and 7 oxygen atoms for both pentanal and hexanal oxidation initiated by OH radicals  
379 within 13 s reaction time. Although the highest precursor concentration (6.4 ppm) was required  
380 for the first observation of detectable HOM in short reaction time experiment with pentanal, a  
381 lower concentration (2.5 ppm) was used to obtain its observed mass spectrum in the long  
382 reaction experiment. The HOM accretion products with up to 11 oxygen atoms were observed  
383 in C<sub>6</sub>–C<sub>8</sub> *n*-aldehyde oxidation experiments while they were limited to maximum 10 oxygen  
384 atoms in pentanal case. We also observed the trend of increased oxidation product yields with  
385 the increase of carbon chain length. In all studied systems, the dominant product signals are O<sub>6</sub>  
386 HOMs with the O<sub>5</sub> HOMs being the second dominant ones. Previous mechanistic  
387 understanding (Barua et al., 2023) as well as current experimental observations reveal that  
388 autoxidation process forming O<sub>5</sub> RO<sub>2</sub> in *n*-aldehydes is very fast while the subsequent  
389 unimolecular rearrangements of the RO<sub>2</sub> intermediates are in competition with bimolecular  
390 reactions including other RO<sub>2</sub>, HO<sub>2</sub>, and NO<sub>x</sub>. The experiments in the presence of high NO  
391 concentrations (30 ppb and above) produced the highest yields of HOM-ONs, compared to  
392 neighboring non-nitrogen HOMs, with the expense of HOM accretion products. However,  
393 some enhancements with the yields of low oxygenated closed-shell products and O<sub>6</sub> peroxy  
394 radicals were also seen under 1–70 ppb NO conditions. The results of hydrogen to deuterium  
395 (H/D) exchange experiments with identical mass shifts in the oxidation products of all studied  
396 *n*-aldehydes imply that the autoxidation mechanism established for hexanal (Barua et al., 2023)  
397 is valid for other *n*-aldehydes. Therefore, accounting for linear aldehydes and their atmospheric  
398 oxidation with increasing importance to longer carbon chain length as a direct source of  
399 condensable materials even under moderately polluted urban areas is essential.

#### 400 ■ DATA AVAILABILITY

401 Details about the experimental setup and mass spectrometry, chemicals and gas cylinders,  
402 mechanistic details of the oxidation steps, additional insights into abundance of HOMs, and  
403 details of kinetic simulations are provided in the Supplement. An Excel file (.xlsx) with results  
404 from mass spectrometry, and two example Kinetiscope (.rxn) files containing kinetic  
405 simulation input parameters—one with NO, another without—are available online  
406 (<https://doi.org/10.5281/zenodo.18894230>).

407    **■ AUTHOR CONTRIBUTIONS**

408    Conceptualization: MR, SB, AK; data curation: SB, AK; formal analysis: SB; investigation:  
409    SB, AK, PS, SI, MR; methodology: SB, AK; writing (original draft preparation): SB; writing  
410    (review and editing): SB, AK, PS, SI, MR; funding acquisition: SI, MR.

411    **■ COMPETING INTERESTS**

412    The authors declare that they have no conflict of interest.

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