Responses to all reviewers:

We thank the reviewers for the careful review of our manuscript. The comments and suggestions are greatly appreciated and have helped to further improve the manuscript. As shown below, all the comments have been addressed. In the following, please find our responses to the comments and the corresponding revisions made to the manuscript. The point-by-point comments are addressed in the following text. The original comments are shown in italics and responses are in normal font. The revised parts of the manuscript are in blue. The line numbers here refer to the clean version.

Reviewer: 1

This manuscript reports high–time/mass resolution online molecular measurements of OA using EESI-TOF at a mountain site in SE China, comparing cloud droplets, interstitial, and cloud-free aerosol. It is clearly within ACP's scope, addressing atmospheric composition and processes with implications for aerosol–cloud interactions and climate. However, the discussion of "tracers" requires more caution: the manuscript should clearly distinguish between true tracers and compounds showing enhanced signals, define the concept explicitly as illustrated below.

Major comments:

1 . Introduction: the current motivation for a cloud-droplet study is underdeveloped. The Introduction should more clearly articulate (i) why in-cloud processing matters for OA burdens and properties, (ii) what key uncertainties remain despite prior fog/cloud and online/offline molecular studies, and (iii) why an online, molecular-level approach (EESI-TOF) is uniquely positioned to address those gaps. Please sharpen the problem statement, specify the hypotheses, and state the concrete research questions and expected contributions of this dataset.

Response: Accepted.

(i)As suggested, in the revise manuscript we have further sharpen the problem statement and added more illustrations on the motivation of our cloud-droplet study in line 56-63 in the manuscript as follows:

"In contrast to fog, cloud is more common, ubiquitously presents in the atmosphere, and consists of a large quantity of droplets generated by aerosol activation, providing an aqueous medium for physical processes and chemical reactions (McNeill et al., 2012; McNeill, 2015). Within clouds, aerosol particles may undergo repeated hydration-dehydration cycles, including hygroscopic growth, activation, and subsequent evaporation. Such cloud processing could influence the concentration of OA composition (Wang et al., 2024; Gao et al., 2023; Liu et al., 2023), thereby influencing aerosol size distribution, hygroscopicity, volatility, and cloud condensation nuclei (CCN) activity (Jimenez et al., 2009; Sun et al., 2024a; Huang et al., 2018).

30 Additionally, cloud processing may facilitate the formation of brown carbon, including N-containing heterocyclic compounds, which could affect atmospheric radiative forcing (Liu et al., 2023)."

*(ii)*Key uncertainties in previous studies, and the necessity of the online molecular-level approach (EESI-TOF) to address these gaps are illustrated in line 64-89 in the manuscript as follows:

"A number of field campaigns have been conducted to measure the chemical composition of OA in cloud droplets. Several previous field campaigns found that more highly oxygenated OA is present in cloud droplets compared to cloud-free aerosol particles using online techniques, Aerosol Mass Spectrometer (AMS) or Aerodyne Aerosol Chemical Speciation Monitor (ACSM), which provide information on fragment ions of compounds, such as the fraction of m/z 44 (CO_2^+) in the mass spectra (Dadashazar et al., 2022; Lance et al., 2020; Gao et al., 2023). Although these studies provide valuable information on the chemical composition of aqSOA, the use of AMS or ACSM leads to molecular fragmentation and thus cannot provide molecular formulas for the components of aqSOA. As a result, the molecular composition of aqSOA and mechanisms of its formation and transformation remain incompletely understood. This gap hinders the analysis of sources, evolution, health effects, and climate impacts with respect to specific OA compounds.

Molecular formulas of OA in cloud samples can be assigned and classified into several groups, including CHO, CHON, CHOS, and CHONS, with CHO and CHON accounting for the largest fractions (Liu et al., 2023b; Cook et al., 2017; Pailler et al., 2024; Zhao et al., 2013; Bianco et al., 2019; Sun et al., 2021; Gramlich et al., 2023). Oligomers (Cook et al., 2017; Zhao et al., 2013), organosulfates (Sun et al., 2021; Bianco et al., 2019), and N-containing compounds such as nitroaromatics (Sun et al., 2021; Cook et al., 2017; Bianco et al., 2019) have been observed in cloud droplets. Although the molecular composition of OA in cloud droplets has been characterized using offline techniques such as Fourier Transform Ion Cyclotron Resonance Mass Spectrometry (FT-ICR-MS), the formation mechanisms of many compounds in clouds remain uncertain. For example, it is not clear whether the oligomers originate from cloud processing or from aqueous aerosol due to a lack of concomitant aerosol measurements and limited temporal variation analyses (Cook et al., 2017; Zhao et al., 2013). The coarse time resolution of filter-based sampling (several hours to one day), together with limited sample numbers, prevents these studies from resolving cloud-processing reaction processes that occur on minute-to-hour timescales and are subjected to the influence of rapid variability in meteorological conditions within clouds. The chemical characteristics of aqSOA obtained from comparisons between cloud droplets and cloud-free aerosol particles are subject to large uncertainties, because the composition of both cloud droplets and aerosols may change over long-time sampling. Therefore, it is necessary to obtain online molecular information on OA in clouds by comparing OA composition of cloud droplets, interstitial aerosol particles, and cloud-free aerosol particles, to provide new insights into the detailed chemical composition, evolution variation, and the mechanism of cloud processing."

2．Line 246-249 and AqSOA tracers about siloxanes. The current discussion and explanation is not accurate. In particular, the concepts of "tracers" and "enhanced compounds" should be clearly distinguished. Please clarify the likely sources and implications of the siloxanes, or restructure this section accordingly. In fact, siloxanes are commonly used in personal care products and industrial chemical products. The lifetimes of the siloxanes are relatively long and will undergo atmospheric oxidation process to generate SOA, as confirmed in both laboratory and ambient studies [1, 2]. But in the aerosol phase, the high-molecular-weight siloxanes have also been detected from vehicle and jet emissions [3, 4]. The siloxanes could also be attributed to the use of silicone tubing in the sampling line [5]. Could the authors confirm that the silicone tubing was not used in the sampling system? It would also be helpful to show the time series of these compounds over the entire campaign. Was there an AMS or PTR-MS available to provide supporting information? Were any siloxane SOA tracers detected (e.g., Si- containing compounds where one or more methyl groups are replaced by hydroxyl groups)? In summary, the discussion could be better connected the focus of this study, as the siloxanes are good surfactants and could influence cloud droplet activation.

Response: Thanks for the reviewer's valuable suggestion. We address the point on "tracers" and "enhanced compounds" in the next response. Regarding siloxanes, we had considered the potential contamination from semi-volatile organic compounds (SVOCs), especially siloxanes, before the campaign. Therefore, stainless steel tubing was primarily used in the sampling line, with copper tubing applied in some sections, and no silicone tubing was used.

As noted by the reviewer, sources of siloxanes are diverse and require further investigation. Discussions on siloxanes are beyond the scope of the present study and will be addressed in a separate manuscript. Consequently, to avoid confusion, we decided to exclude the siloxanes discussion in the revised manuscript, and data points of siloxanes have been removed from Figure 5 and Figure S4 (both shown below).

We have revised line 276-277 in the manuscript as follows:

"In addition, some siloxane compounds showed higher fractions in CD than in CF, such as C16H48O8Si8 (m/z 615) and C18H54O9Si9 (m/z 689) in CE2 and C12H36O6Si6 (m/z 467) and C18H54O9Si9 in CE3. Siloxane is a type of volatile chemical products such as those found in personal care products. To our best knowledge, this is the first time that C16H48O8Si8 was observed in cloud droplets. The reason for the higher fraction of siloxane warrants further study.
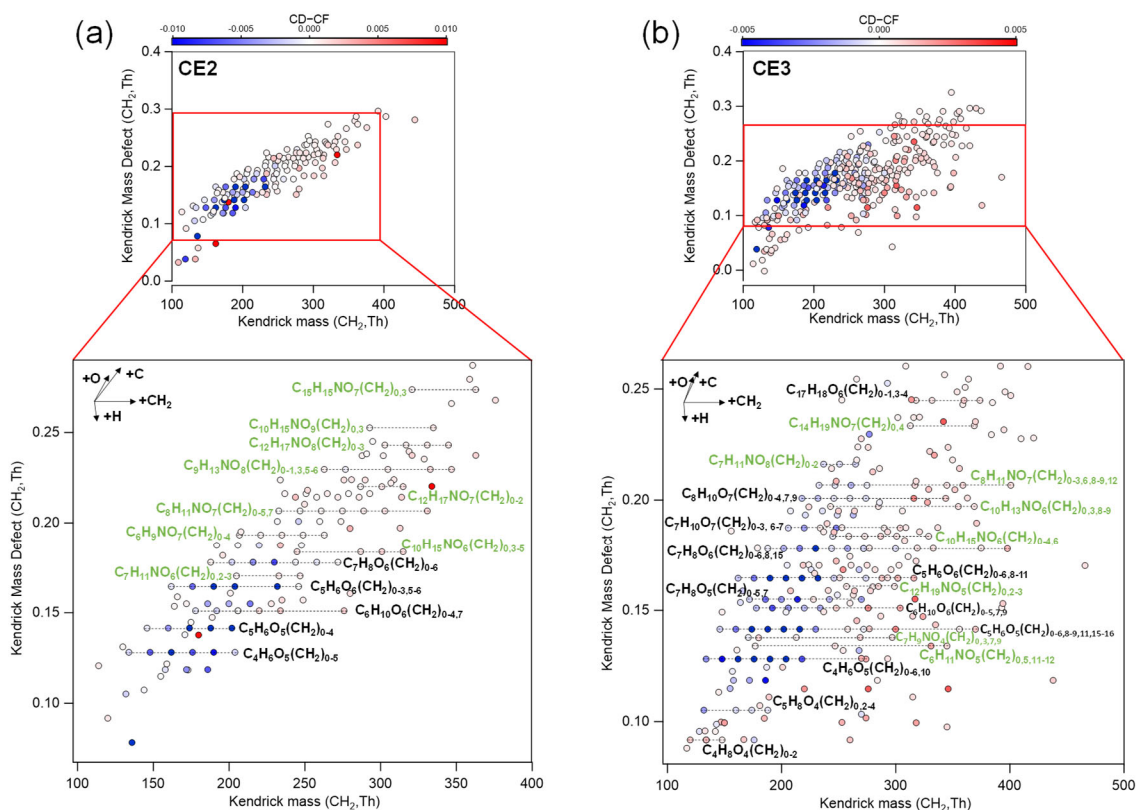
Figure 5. Kendrick mass defect plots based on CH_2 of compounds in (a) CE2 and (b) CE3. Data points are color-coded by differences in fractions of compounds between CD and CF. Fractions of compounds are normalized to the sum of signals of all organics in CD and CF, respectively. Note that, for conciseness, data points in CE3 with normalized signal difference between −0.0003 and 0.0003 (appearing nearly white) are not shown here. Siloxane compounds are not shown here for clarity.
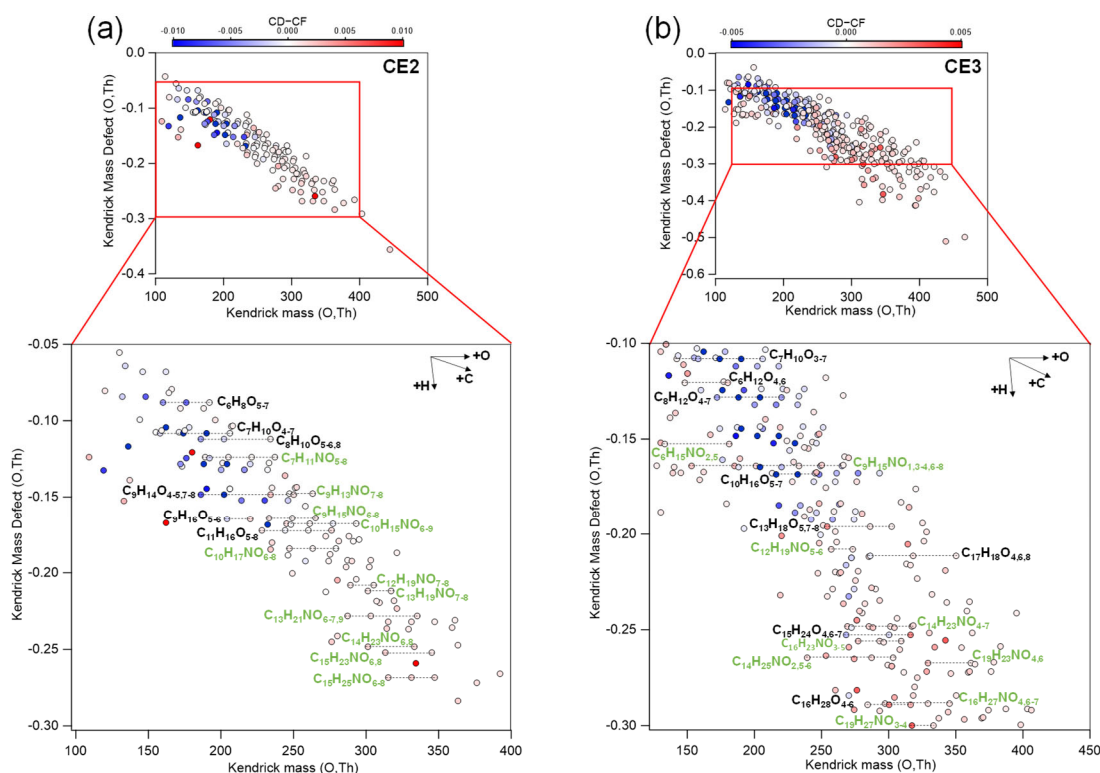

Figure S4. Kendrick mass defect plots based on O of compounds in (a) CE2 and (b) CE3. Data points are color-coded by differences in fraction of compounds between CD and CF. Fraction of compounds are normalized to the sum of signals of all organics in CD and CF, respectively. Note that, for conciseness, data points in CE3 with normalized signal difference between −0.0003 and 0.0003 (appearing nearly white) are not shown here. Siloxane compounds are not shown here for clarity.

Response: Thanks for the reviewer's suggestion. We agree with the reviewer that the terms "aqSOA tracers" we used in the manuscript may cause some misunderstanding. The "39 tracers" in the original manuscript are compounds that are significantly enriched in cloud droplets rather than in cloud-free aerosol particles. In the revised manuscript, we change the terminology to "enriched OA compounds" which is also used in Sun et al. (2024b). The 39 enriched OA compounds are divided into four classes: CHO, CHON, CHN, and CHOSi. The CHN-class OA compounds may be heterocyclic compounds containing imine or amine functional groups, potentially resulting from secondary formation in the aqueous phase. Also, the CHN-class compounds may come from primary sources, arising from the uptake of aerosol-phase organics. $C_9H_{18}N_2$ could be emitted from traditional biomass fuel burning (Fleming et al., 2018) and $C_{12}H_{23}N$ from vehicle emissions (Thomas et al., 2025). And agricultural residue burning could emit $C_9H_{18}N_2$ and $C_{12}H_{23}N$ (Lin et al., 2012). Sources of CHOSi-class compounds warrant further investigation. The CHO, CHON, and CHN-class OA compounds could be potential aqSOA tracers for enrichment in cloud droplets. And molecular structures are warranted in future to verify formation mechanisms.

We have revised line 285-296 in the manuscript as follows:

"We identified aqSOA tracers in cloud droplets by comparing the intensity fractions of all compounds between CD and CF using a *t*-test at a significant level of 0.05. A total of 144, 421, 274, and 537 organic compounds in CE1, CE2, CE3, and CE4, respectively, passed the *t*-test. Among these compounds, 39 organic compounds in CD were significantly enriched in three or four CEs, as shown in Table 2. Two were consistently significant in CD across all four CEs: $C_{14}H_{42}O_7Si_7$ and $C_9H_{22}N_2O_4$. Furthermore, sulfate compounds were enriched in CD compared with CF in three CEs, of which time series is shown in Fig. S6. Sulfate is a well-established tracer for aqueous-phase processing, and its elevated concentration in cloud droplets and fog has been widely reported (Dadashazar et al., 2022; Brege et al., 2018; Kim et al., 2019), which further enhances the potential of identifying the enriched OA compounds as aqSOA tracers formed via cloud processing. The number of CHO, CHON, CHN, and CHOSi compounds is 15, 19, 2, and 3, respectively. The majority of the enriched OA compounds exhibit carbon numbers greater than nine, which is also an indication of accretion reactions in cloud droplets. Most of these enriched

OA compounds have not been reported in previous literature (Cook et al., 2017; Bianco et al., 2019; Sun et al., 2024b; Tong et al., 2021)."

Line 31-32 in Abstract have been revised as follows:

"We identified 39 significantly enriched compounds in CD compared with CF, which could be potentially used as aqSOA tracers formed via cloud processing."

Line 361-363 in Conclusion have been revised as follows:

"We identified several compounds significantly enriched in cloud droplets, including typical aqSOA tracers such as oxalic acid. The new aqSOA tracers, such as $C_6H_{12}O_6$ and $C_9H_{22}N_2O_4$, could help future studies identify cloud processing aqSOA."

4 . The manuscript relies heavily on earlier and in-campaign citations (71 % before 2020) and does not sufficiently reflect recent literature (29% from the most recent 5 years, e.g. Line 48-57, line 219-222, line 246-294). In particular, line 269 lists tracer studies from 2017 and 2019, but more up-to-date work is available from 2019-2025. Note that EESI-TOF was developed around 2017, first field measurements appeared in 2018–2019 (Switzerland), and the first report of aqSOA measured by EESI was published in 2021. The manuscript should be revised to incorporate these and subsequent studies, ensuring comprehensive and current coverage beyond the authors' own campaigns.

Response: Thanks for your kind suggestion. We added more references of aqSOA measured by EESI-ToF-MS in the revised manuscript as follows:

Line 47-63 in Introduction:

"Mounting evidence for aqueous secondary organic aerosol (aqSOA) has been reported in field observations in various atmospheric aqueous systems, i.e., aerosol liquid water (ALW), fog water, and cloud water. For example, several studies on source apportionment in different sites showed that aqSOA formed in ALW is an important contributor to SOA, with its fraction particularly elevated (up to 44 %) under high relative humidity (RH) conditions (Wang et al., 2021; Zhao et al., 2019; Tong et al., 2021; Gilardoni et al., 2016; Duan et al., 2022; Xu et al., 2019; Sun et al., 2016). Relative to ALW, fog water and cloud water are diluted aqueous systems where aqSOA can also be formed (Herckes et al., 2013). For fog water, the ratio of aqSOA to OA during fog-rain days is enhanced compared with non-fog-rain days (Duan et al., 2021). Additionally, OA composition of fog water is more oxidized (Brege et al., 2018), has more N-containing compounds (Mattsson et al., 2025; Sun et al., 2024a; Kim et al., 2019) compared with aerosol particles, and shows signs of oligomerization based on fragments in the mass spectrum (Gilardoni et al., 2016; Mandariya et al., 2019). In contrast to fog, the cloud is more common, ubiquitously presents in the atmosphere, and consists of a large quantity of droplets generated by aerosol activation, providing an aqueous medium for physical processes and chemical reactions (McNeill et al., 2012; McNeill, 2015). Within clouds, aerosol particles

may undergo repeated hydration-dehydration cycles, including hygroscopic growth, activation, and subsequent evaporation. Such cloud processing could influence the concentration of OA composition (Wang et al., 2024b; Gao et al., 2023; Liu et al., 2023b), thereby influencing aerosol size distribution, hygroscopicity, volatility, and cloud condensation nuclei (CCN) activity (Jimenez et al., 2009). Additionally, cloud processing may facilitate the formation of brown carbon, including N-containing

160   heterocyclic compounds, which could affect atmospheric radiative forcing (Liu et al., 2023b)."

Line 120-121 in Methods:

"Detailed information regarding EESI-ToF-MS has been reported previously (Lopez-Hilfiker et al., 2019; Stefenelli et al., 2019; Brown et al., 2021; Kumar et al., 2022; Luo et al., 2024; Xue et al., 2025)."

Line 238-248 in Sect. 3.2:

165   "Collectively, compounds in CD had more n_C, n_O, and n_N than those in CF. The molecular characteristic of higher n_C is likely attributed to accretion reactions such as oligomerization (Yu et al., 2016; Fenselau et al., 2025). This finding is consistent with several laboratory studies of aqSOA formation. For instance, enriched high-molecular-weight compounds (HMWC) in aqSOA were reported in the bulk phase experiments of methylglyoxal and glyoxal under cloud-relevant conditions (Tan et al., 2009; Altieri et al., 2008). And aqSOA from in-cloud simulation using a wetted-wall flow reactor has more highly oxygenated

170   and carbon-containing compounds than gasSOA simulated by an oxidation flow reactor (OFR) from the same biomass burning samples (Wang et al., 2024b). Experiments in the bulk phase and the wetted-wall flow reactor which better represents atmospheric aqueous conditions, indicate that accretion reactions could be prevalent in cloud droplets. Field observations in the Arctic also show potential evidence of accretion reactions, with compounds of longer carbon chains enriched in CD relative to CF (Pasquier et al., 2022), hinting at the possible importance of accretion reactions. Notably, this study provides direct

175   molecular-level evidence for the contribution of accretion reactions during cloud processing of OA."

Line 292-296 in Sect. 3.3:

"The number of CHO, CHON, CHN, and CHOSi compounds is 15, 19, 2, and 3, respectively. The majority of the enriched OA compounds exhibit carbon numbers greater than nine, which is also an indication of accretion reactions in cloud droplets. Most of these enriched OA compounds have not been reported in previous literature (Cook et al., 2017; Bianco et al., 2019;

180   Tong et al., 2021; Sun et al., 2024b).

Minor comments:

Line 61: "Aerosol Mass Spectrometer (AMS) or Aerodyne Aerosol Chemical Speciation Monitor (ACSM)".

Response: We thank the reviewer for the kind remarks. We have revised line 64-68 in the manuscript as follows:

185   "A number of field campaigns have been conducted to measure chemical composition of OA in cloud droplets. Several previous field campaigns found that more highly oxygenated OA was present in cloud droplets compared to cloud-free aerosol particles using online techniques, Aerosol Mass Spectrometer (AMS) or Aerodyne Aerosol Chemical Speciation Monitor (ACSM), which obtained information on fragment ions of compounds, such as the fraction of m/z 44 (CO_2^+) in the mass spectra (Dadashazar et al., 2022; Lance et al., 2020; Gao et al., 2023)."

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Line 82: revise to "...at the summit of Damaojian Mountain, located in Jinhua City, Zhejiang Province, China."

Response: We have revised line 97-99 in the manuscript as follows:

    "We conducted this field campaign from May 1st to May 29th in 2024 at Shanghuang Eco-Environmental Observatory of Chinese Academy of Sciences at the summit of the Damaojian mountain (119.51° E and 28.58° N, 1128 m above sea level)

195 ~~that is~~ located in Jinhua city, Zhejiang province, China."

Line 105: delete "Here is a brief introduction.", too informal.

Response: We have revised line 119-124 in the manuscript as follows:

    "Detailed information regarding EESI-ToF-MS has been reported previously (Lopez-Hilfiker et al., 2019; Stefenelli et

200 al., 2019; Brown et al., 2021; Kumar et al., 2022; Luo et al., 2024; Xue et al., 2025). ~~Here is a brief introduction.~~ Aerosol was sampled after gaseous compounds were removed by entering a charcoal denuder, and subsequently intersected with an electrospray generated from a working solution containing 100 ppm NaI in a 1:1 (v/v) water and acetonitrile mixture, allowing aerosol compounds to be detected as [M+Na]$^+$ in positive ion mode."

205 *Line 115: "signal-to-background ration (s/b)".*

Response: We have revised line 129-132 in the manuscript as follows:

    "Mass spectral data were processed using Tofware 3.2.5 in Igor Pro 8. For data screening, the signal-to-background ratio (s/b) was calculated as the median value of (sample signal−background)/background, thereby excluding compounds showing insignificant differences between sample and background."

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Line 268: "To the best of our knowledge, this is the first observation of C16H48O8Si8 ...". As mentioned in major comments 2, the description needs to be considered.

Response: We have deleted this sentence based on the response to major comment#2 in the revised manuscript.

215 *Line 308: hours to a day.*

Response: This has been modified in line 343-345 in the revised manuscript as follows:

"Most previous comparisons of the chemical composition of cloud droplets with cloud-free aerosol particles or interstitial aerosol particles are based on long sampling (hours to a day) and offline analysis (Sun et al., 2021; Liu et al., 2023b)."

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1. *Chen, Y., et al., Chemical characterization and formation of secondary organosiloxane aerosol (SOSiA) from OH oxidation of decamethylcyclopentasiloxane. Environmental Science: Atmospheres, 2023. 3(4): p. 662-671.*

2. *Meepage, J.N., et al., Advances in the Separation and Detection of Secondary Organic Aerosol Produced by Decamethylcyclopentasiloxane (D5) in Laboratory-Generated and Ambient Aerosol. ACS ES&T Air, 2024. 1(5): p.*

230   *365-375.*

3. *Yao, P., et al., Methylsiloxanes from Vehicle Emissions Detected in Aerosol Particles. Environmental Science & Technology, 2023. 57(38): p. 14269-14279.*

4. *Decker, Z.C.J., et al., Emission and Formation of Aircraft Engine Oil Ultrafine Particles. ACS ES&T Air, 2024. 1(12): p. 1662-1672.*

235 5. *Timko, M.T., et al., Composition and Sources of the Organic Particle Emissions from Aircraft Engines. Aerosol Science and Technology, 2014. 48(1): p. 61-73.*

240 **Reference：**

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