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: 2

The study characterized the molecular composition of the organic fraction of cloud droplet residue, pre-cloud aerosols, interstitial aerosols, and post-cloud aerosols using EESI-ToF-MS. The approach yielded a high-resolution dataset that captured detailed temporal variations in molecular composition. While the study provided additional evidence on the relative contributions of CHO and CHON species in cloud droplet residues and aerosols, its findings largely corroborated previous results rather than extending current understanding. The authors interpreted their data mostly relying on findings from previous studies.

Response: We thank the reviewer for the comments on our manuscript. While we appreciate the reviewer's perspective, we respectfully disagree with the comment that our "findings largely corroborated previous results rather than extending current understanding". Here are three new findings in our study that extend previous understanding.

1. We find that the CHON compounds enhanced in cloud droplets compared to interstitial and cloud-free aerosol particles are mostly contributed by compounds with an O/N ratio of $\geqslant 3$, likely organonitrates, amino acids, or nitrogen-heterocyclic compounds. This finding is in contrast to previous studies. E.g., Liu et al. (2023) reported that the number of CHON compounds was higher in cloud droplets in Mt. Tai using FT-ICR-MS, and more CHON compounds had reduced nitrogen groups (O/N <3). Our results indicate different formation mechanisms of CHON compounds.
2. We provide direct molecular-level evidence for the contribution of accretion reactions during cloud processing of OA. Fog samples measured by AMS were compared with aerosol at different stages of fog episodes, suggesting that oligomerization could be significant in fog processing of OA (Mandariya et al., 2019). However, due to the fragmentation in AMS, there was no direct evidence. Although cloud observations using FT-ICR-MS have reported the presence of oligomers in cloud samples, such studies could not determine the source of oligomers, whether they originated from cloud processing or from uptake of gas-phase or aqueous aerosol, because no concomitant aerosol samples were collected for comparison (Zhao et al., 2013; Cook et al., 2017). In this study, by comparing OA composition in cloud droplets with that in cloud-free aerosol particles, we provide molecular-level evidence from cloud observations and prove that accretion reactions

have a substantial contribution to aqSOA.

3. We find several enriched compounds in cloud droplets compared to cloud-free aerosol particles. Most of the enriched OA compounds have not been reported in previous literature (Cook et al., 2017; Bianco et al., 2019; Tong et al., 2021; Sun et al., 2024b), and could possibly be used as tracers of cloud-processing aqSOA. This can be helpful for further study to identify the contribution of cloud processing.

In addition, previous offline studies are limited by low temporal resolution and limited sample numbers (Sun et al., 2021; Cook et al., 2017; Pailler et al., 2024), whereas existing online measurements provided only fragmented OA information without molecular formulas (Dadashazar et al., 2022; Lance et al., 2020). By combining 20-s time resolution with molecular-level characterization from EESI-ToF-MS, this study tracks the formation and evolution of aqSOA during cloud processing and offers new insights into cloud-influenced organic aerosol chemistry.

We thank the reviewer for the comments. We realize that some descriptions in the original manuscript were not sufficiently clear to convey our intended findings, which may have led to misunderstanding. To address this, we have strengthened the explanation of the novelty and clarified the related statements in the introduction, discussion, and conclusion of the revised manuscript as follows:

Line 64-89 in Introduction:

"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., 2023; 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."

Line 245-248 in Sect 3.2:

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

Line 352-389 in Conclusion:

"AqSOA molecular composition and processing in cloud episodes were studied using online molecular information obtained by EESI-ToF-MS at a high-mountain site in China. Cloud processing substantially influences OA composition, resulting in large differences among distinct cloud episodes. Organics in cloud droplets had an average molecular formula $C_{9.95–12.92}H_{14.53–21.78}O_{5.15–6.02}N_{0.32–0.42}S_{0–0.01}Si_{0–1.29}$ for the selected four cloud episodes. CHO compounds contributed predominantly to OA in cloud droplets. CHON was enhanced markedly in cloud droplets compared with cloud-free aerosol particles and interstitial aerosol particles in most cloud episodes. The majority of CHON compounds were likely organonitrates, highlighting the enrichment of organonitrates compounds in cloud processing. OA in cloud droplets contained higher numbers of C, O, and N atoms, exhibited a CH_2-based homologous series, and showed an enrichment of higher-molecular-weight compounds compared with adjacent cloud-free aerosol particles, collectively highlighting the importance of accretion reactions in cloud processing of OA at the molecular level. 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.

This study provides direct molecular-level evidence for the contribution of accretion reactions during cloud processing of OA. Although previous cloud observations using FT-ICR-MS reported the presence of oligomers in cloud samples, these studies could not distinguish whether such compounds originated from cloud processing or aqueous aerosols, as no concomitant aerosol samples were collected for comparison (Zhao et al., 2013; Cook et al., 2017). By directly comparing OA composition in cloud droplets with that in cloud-free aerosol particles, our results clearly demonstrate that accretion reactions occur within cloud droplets. It has been assumed that HMWC are predominantly formed in aerosol liquid water rather than cloud water, owing to the lower reaction rates of accretion reactions in the more dilute cloud-water environment (Ervens et al., 2011). In contrast, our study provides direct molecular-level evidence that such compounds can also be formed in cloud water, extending earlier observations by Cook et al. (2017). These findings highlight that accretion reactions should be considered when modeling aqSOA formation in clouds.

The HMWC formed via accretion reaction may have implications for the environment and climate. Due to the increase in the HMWC, accretion reactions likely reduce the volatility of organics and could potentially enhance OA mass concentration and alter the aerosol size distribution after cloud evaporation. The formation of HMWC can also modify physicochemical properties, such as lifetime, oxidation state, viscosity, and hygroscopic properties, which may further influence the cloud activation of these aerosols. In addition, the formation of N-containing compounds in cloud droplets, such as organonitrates, pyrrole, and imidazole, may also affect the physicochemical properties of aqSOA, e.g., contributing to brown carbon and thus affecting regional radiative forcing.

Based on the measurement of high time resolution (~20 s), we find that the concentrations of individual organic compounds were highly dynamic in clouds, which is likely due to the turbulence in clouds. Such a highly dynamic nature in clouds poses difficulties in extracting the influence of chemical processes on individual compounds for instrumentation with low temporal resolution. Therefore, our results highlight the necessity of high time resolution measurements (< 1 h), especially online systems achieving minute-level resolution to investigate the chemical processes in clouds, considering dynamic variations of compounds in clouds due to turbulence in clouds and alterations in air masses.

It should be noted that this study provides molecular formulas only, while detailed structural information is warranted to better constrain the sources, formation mechanisms, and climate impacts of aqSOA in clouds. In addition, sources of compounds enriched in cloud droplets will be investigated in future studies."

The study's main points were: 1. 39 new compounds in cloud droplet residues; and 2. the hypothesis that accretion reactions were promoted during cloud processing. However, it remains unclear why these new 39 compounds were detected. Were they revealed because of the dataset's high temporal resolution, the unique analytical technique, or chemistry specific to the sampled environment?

Response: The "39 compounds" enriched in cloud droplets were detected owing to the high temporal resolution of our measurements and the molecular-level comparison between cloud droplets and cloud-free aerosol particles. The enhancement of OA compounds in cloud droplets could be blurred by methods with low temporal resolution. Additionally, the observation site is subjected to diverse influences, including both anthropogenic and biogenic contributions, where aqueous processing of various compounds can be identified. We would like to clarify that the "39 new compounds" refer to newly identified aqSOA tracers that expand the existing reservoir of known tracers. In fact, some of these compounds have been reported in previous aerosol studies (e.g., $C_{10}H_{16}O_2$ in Qi et al. (2019), but their enrichment in cloud droplets and their role as aqSOA tracers have not been recognized before.

And following Reviewer 1's suggestion, we have modified the discussion of the 39 enriched compounds in line 285-296 in Sect. 3.3 of the revised manuscript to clarify their definition 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., 2024; Tong et al., 2021)."

The study period experienced four cloud events, effectively providing four data points for comparing pre-cloud, CD, INT and CF samples. Caution should be exercised in drawing general conclusions from such a limited dataset. For instance, CE2 was an exception in which CHON did not comprise the largest fraction of total OA, and CHO was not the lowest among the four sample types. Thus, one quarter of the samples did not align with the generalization, and it would be advisable to moderate some of the claims derived from these comparisons.

Response: Thanks for the reviewer's reminder and comments. CE2 shows an exception of CHO and CHON characteristics from other CEs due to air mass changes during the long-time interval between post-cloud and others (pre-cloud, CD, and INT). To make the statements more accurate, we have revised line 188-191 in the manuscript as follows:

"In most cloud episodes, CD showed the lowest CHO fraction (54.8 %–70.7 %) and the highest CHON fraction (26.6 %–33.2 %) among the four sample types. ~~The only exception i~~In CE2, with higher CHON (29.4 %) and lower CHO (54.6 %) in post-cloud aerosols than other sample types, is attributable to air mass changes during the long-time interval between post-cloud and others (shown in Fig. 2 and Fig. S1), as indicated by elevated CO concentration."

Line 28-30 in Abstract are revised as follows:

"In most cloud episodes, the fraction of CHO was lower in CD than that in INT and CF, while the fraction of CHON was higher, which may result from the uptake of organonitrates or nitration in cloud water."

Line 356-357 in Conclusion are revised as follows:

"CHON was enhanced markedly in cloud droplets compared with cloud-free aerosol particles and interstitial aerosol particles in most cloud episodes."

Specific comments:

1. Line 27: "With adjacent time" is not clear. Adjacent to what?

Response: We intended to express that the cloud-free aerosol particles were sampled immediately before or after each in-cloud stage, ensuring that the comparison among cloud droplets, interstitial aerosol particles, and cloud-free aerosol particles was conducted under similar meteorological and emission conditions. Therefore, the differences observed in OA composition can be attributed primarily to cloud processing rather than temporal variability. To avoid ambiguity, the phrase "with adjacent time" has been removed, and the expression has been revised for clarity in the manuscript.

Line 26-28 in the revised manuscript have been modified as follows:

"We identified 2084 molecular formulas and compared OA composition from three sample types ~~with adjacent time (<2 h)~~: cloud droplets (CD), interstitial aerosol particles (INT), and cloud-free aerosol particles (CF) in representative cloud episodes."

2. Line 44-46: Research on the role of aqueous-phase chemistry in SOAs has been going on for more than four decades. Are the studies cited here seminal ones?

Response: Thank you for the suggestion. We have gone through the literature on the role of aqueous-phase chemistry in SOA and added references in line 45-46 in Introduction as follows:

“In addition to the traditional gas-phase processing, aqueous-phase pathways have been recognized as an important source of SOA (Sehested et al., 1975; Galloway et al., 1976; Graedel and Weschler, 1981; Fu et al., 2008; Tan et al., 2009; Zhang et al., 2010; Ervens et al., 2011; Lamkaddam et al., 2021).”

3. Line 51-52: What are the main findings from Duan et al. (2021)?

Response: Thanks for the reminder. We have expanded the main findings of Duan et al. (2021) in Line 52-55 in the revised manuscript as follows:

“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).”

4. Lines 151-153: Are the ranges for C, H, O, and N – C:10.01–12.81 vs 8.43–11.10, H: 14.59–20.34 vs. 14.23–16.83, O: 5.08–6.00 vs. 5.06–5.72, and N: 0.34–0.43 vs. 0.16–0.35 – between CD and pre-loud really that different?

Response: We have done statistics tests, which show that the numbers of C, H, O, and N atoms of organic compounds in CD are indeed significantly higher compared to pre-cloud ($p<0.05$). In addition, we have updated these values according to an optimized method in which raw signal values at each time in CD and pre-cloud are used to calculate the average molecular formula, while average signal values in the whole CD and pre-cloud periods were used in the original manuscript. The increase in n_C suggests the occurrence of accretion reactions in cloud droplets, leading to the formation of higher-molecular-weight compounds. The enhancement in n_O indicates a higher degree of oxidation in cloud droplets. The increase in n_N is consistent with the results discussed in Sect. 3.2, where we demonstrated that CHON compounds are enriched in cloud droplets relative to cloud-free aerosol particles.

Line 167-178 in Sect. 3.1 are revised as follows:

“A total of 2084 molecular formulas of OA were identified in the campaign. Mean formula of CD was $C_{9.95–12.92}H_{14.53–21.78}O_{5.15–6.02}N_{0.32–0.42}S_{0–0.01}Si_{0–1.29}$ for CE1–CE4. Compared with pre-cloud aerosols with formula $C_{8.17–10.57}H_{12.99–16.14}O_{4.98–5.64}N_{0.12–0.30}S_{0–0.01}Si_{0–0.24}$, CD exhibited increased numbers of carbon, hydrogen, oxygen, and nitrogen atoms, with the differences being statistically significant ($p< 0.05$) (Table S2). These molecular formulas were classified into eight classes, that is, CHO (only C, H, O atoms are contained in the chemical formula, hereafter), CHON, CHONS, CHOS, CHN, CHS, CHNS, and CHOSi. Since the composition of OA varied in different CEs, the fractions of these OA classes are discussed for each CE in Sect. 3.2. The O/C ratio was generally lower in CD (0.45–0.66) than in pre-cloud aerosols, INT, and post-cloud aerosols in 4 CEs. The O/C ratio in CD is comparable to those reported for fog water (0.52–0.68), aqSOA (0.61–0.84), and oxygenated OA (0.44–0.83) by Gilardoni et al. (2016). In general, O/C of CD in this study is comparable to that of fog (0.58–0.8) in the Po Valley in Brege et al. (2018), while H/C of CD (1.50–1.87) is higher than that of fog (1.29–1.37) in that study. Furthermore, CD showed elevated N/C (0.033–0.045) relative to other sample types, while its OSc value (−0.83 to −0.25) is generally lower than in other sample types."

We have added the average atom numbers of C, H, O, and N with standard deviation in Table S2 in the revised supplementary as follows:

Table S2: Mean atom numbers (n_C, n_H, n_O, and n_N; mean ± standard deviation) of OA for CE1–CE4 in pre-cloud aerosols, CD, INT, and post-cloud aerosols.

		Pre-cloud aerosols	CD	INT	Post-cloud aerosols
CE1	n_C	8.72±0.38	10.45±1.07	8.99±0.53	8.86±0.41
	n_H	13.81±1.09	17.25±3.16	14.21±1.17	14.45±1.10
	n_O	5.64±0.12	6.02±0.33	5.68±0.13	5.71±0.15
	n_N	0.20±0.03	0.32±0.08	0.26±0.05	0.22±0.04
CE2	n_C	8.33±0.20	10.13±1.26	8.52±0.34	11.12±0.55
	n_H	13.72±0.65	19.51±4.50	14.19±1.05	21.10±2.80
	n_O	4.98±0.12	5.44±0.57	4.92±0.18	6.26±0.20
	n_N	0.28±0.04	0.39±0.09	0.33±0.07	0.33±0.07
CE3	n_C	10.57±0.18	12.92±0.70	10.42±0.09	10.51±0.13
	n_H	16.14±0.30	21.78±1.76	16.21±0.20	16.18±0.29
	n_O	5.08±0.05	5.15±0.12	5.09±0.03	5.11±0.04
	n_N	0.30±0.02	0.42±0.06	0.27±0.02	0.27±0.02
CE4	n_C	8.17±0.15	9.95±0.49	8.94±0.20	8.77±0.12
	n_H	12.99±0.19	14.53±0.65	13.36±0.26	13.20±0.17
	n_O	5.29±0.08	5.68±0.23	5.52±0.08	5.52±0.06
	n_N	0.12±0.03	0.32±0.08	0.20±0.04	0.18±0.02

Table 1 is revised as follows:

Table 1. Mean molecular formulas and elemental parameters (H/C, O/C, N/C, and OSc; mean ± standard deviation) of OA for CE1–CE4 in pre-cloud aerosols, CD, INT, and post-cloud aerosols.

		Pre-cloud aerosols	CD	INT	Post-cloud aerosols
CE1	Formula	$C_{8.72}H_{13.81}O_{5.64}N_{0.20}S_{0}Si_{0.24}$	$C_{10.45}H_{17.25}O_{6.02}N_{0.32}S_{0}Si_{0.69}$	$C_{8.99}H_{14.21}O_{5.68}N_{0.26}S_{0}Si_{0.23}$	$C_{8.86}H_{14.45}O_{5.71}N_{0.22}S_{0}Si_{0.40}$
	H/C	1.61±0.04	1.64±0.11	1.61±0.04	1.61±0.04
	O/C	0.71±0.02	0.66±0.03	0.69±0.02	0.70±0.02
	N/C	0.022±0.004	0.033±0.008	0.028±0.005	0.025±0.006
	OSc	-0.19±0.06	-0.33±0.14	-0.22±0.06	-0.20±0.06
CE2	Formula	$C_{8.33}H_{13.72}O_{4.98}N_{0.28}S_{0.01}Si_{0.16}$	$C_{10.13}H_{19.51}O_{5.44}N_{0.39}S_{0.01}Si_{1.29}$	$C_{8.52}H_{14.19}O_{4.92}N_{0.33}S_{0.01}Si_{0.20}$	$C_{11.12}H_{21.10}O_{6.26}N_{0.33}S_{0.01}Si_{1.52}$
	H/C	1.70±0.03	1.87±0.15	1.72±0.04	1.78±0.13
	O/C	0.63±0.01	0.58±0.03	0.61±0.02	0.61±0.02
	N/C	0.037±0.004	0.045±0.01	0.042±0.009	0.031±0.006
	OSc	-0.43±0.04	-0.71±0.19	-0.50±0.08	-0.56±0.15
CE3	Formula	$C_{10.57}H_{16.14}O_{5.08}N_{0.30}S_{0.01}Si_{0.02}$	$C_{12.92}H_{21.78}O_{5.15}N_{0.42}S_{0.01}Si_{0.87}$	$C_{10.42}H_{16.21}O_{5.09}N_{0.27}S_{0.004}Si_{0.11}$	$C_{10.51}H_{16.18}O_{5.11}N_{0.27}S_{0.01}Si_{0.07}$
	H/C	1.57±0.01	1.73±0.08	1.59±0.01	1.58±0.01
	O/C	0.55±0.008	0.45±0.03	0.55±0.005	0.55±0.005
	N/C	0.033±0.002	0.040±0.007	0.028±0.002	0.029±0.002
	OSc	-0.48±0.02	-0.83±0.12	-0.49±0.02	-0.47±0.02
CE4	Formula	$C_{8.17}H_{12.99}O_{5.29}N_{0.12}S_{0.004}Si_{0}$	$C_{9.95}H_{14.53}O_{5.68}N_{0.32}S_{0.01}Si_{0}$	$C_{8.94}H_{13.36}O_{5.52}N_{0.20}S_{0.004}Si_{0}$	$C_{8.77}H_{13.20}O_{5.52}N_{0.18}S_{0.004}Si_{0}$
	H/C	1.65±0.02	1.50±0.04	1.52±0.02	1.52±0.01
	O/C	0.69±0.01	0.62±0.02	0.66±0.008	0.67±0.01
	N/C	0.014±0.005	0.034±0.01	0.021±0.005	0.019±0.003
	OSc	-0.28±0.04	-0.25±0.07	-0.20±0.02	-0.18±0.02

5. *Line 278: "Large fluctuations" in what?*

Response: We meant large inhomogeneity within clouds due to strong turbulence. We have revised the sentence in Line 305-306 as follows:

"The oxalic acid signal was exclusively observed during CD, whereas it remained weak and noisy in CF and INT, as shown in Fig. 6. The oxalic acid signal was significantly enhanced only in CE2 and CE4, rather than in all CEs, which may be related to larger inhomogeneity within clouds due to strong turbulence."

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