



Supplement of

Molecular composition and processing of aqueous secondary organic aerosol in clouds at a mountain site in southeastern China

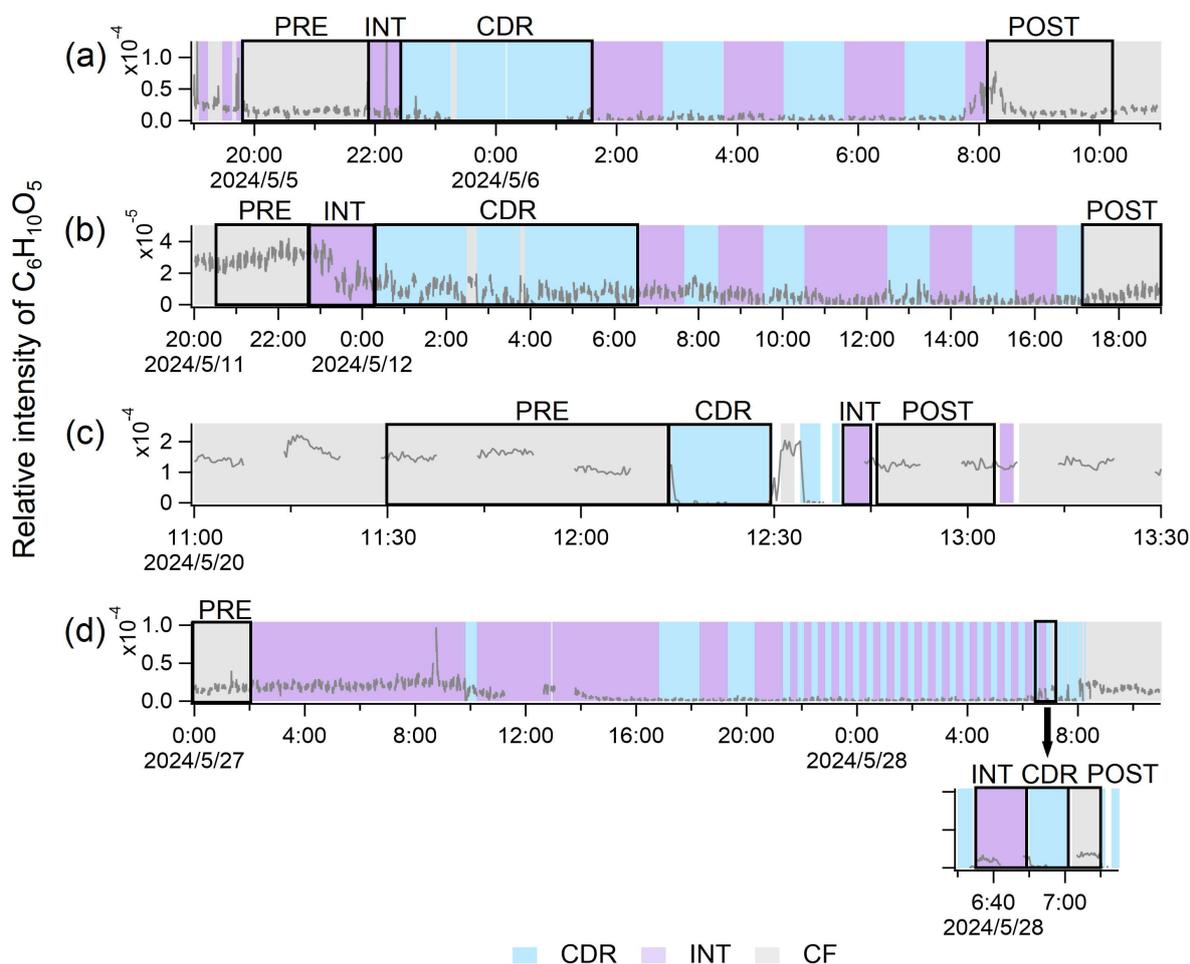
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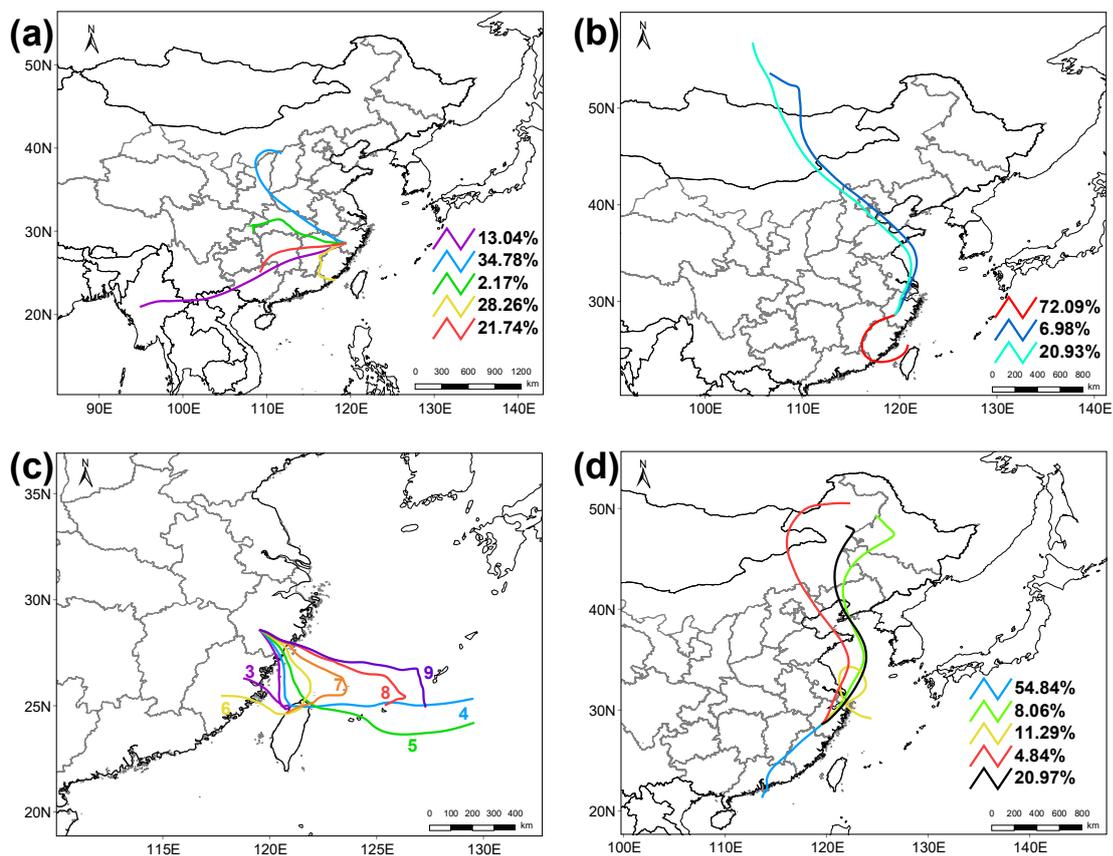
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Text S1. Meteorological condition and backward trajectories

10 The temperature (T) of CE1 was the lowest among the four CEs, while the T of CE3 was the highest. In CE2, RH was
the highest in four CEs. The air mass of CE1 originated from the southwest of the Shanghuang site. The duration time of CE2
was 22.5 hours, during which the origin of air mass turned from southwest (Fujian and Jiangxi provinces) to north (Mongolia)
at 12:00 on May 12th, resulting in a marked decrease in PM_{2.5} concentration. CE3 lasted for 1.5 h, the shortest duration time,
so RH and T were more stable. The air mass in CE3 originated from the ocean in the southeast, which is different from other
15 CEs. The duration time of CE4 is 34 h, the longest of all CEs, the air mass of which came from the southwest and turned to
the northeast in the second half of the duration time at 17:00 on May 27th.

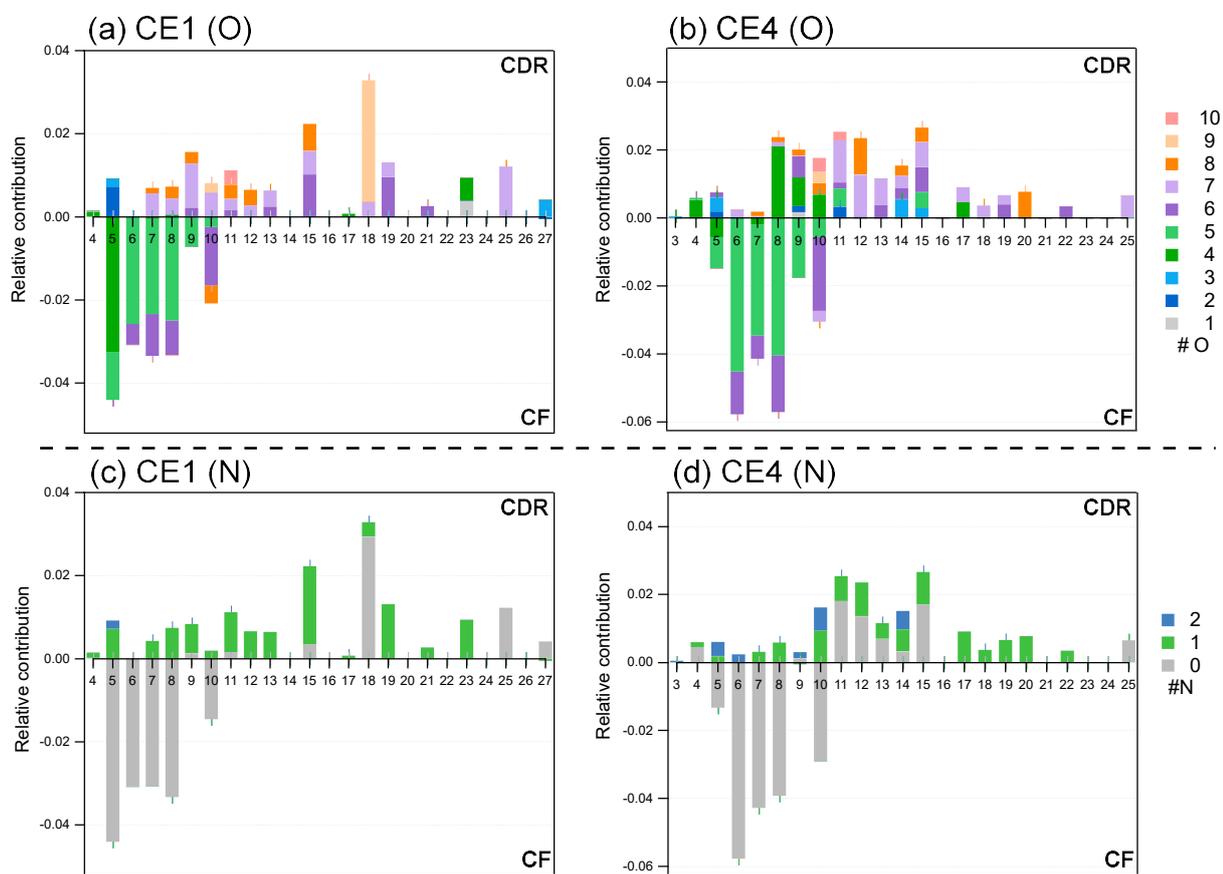


20 **Figure S1. Operational definition of the pre-cloud, in-cloud (including sample types of CDR and INT) and post-cloud stages in (a) CE1, (b) CE2, (c) CE3, and (d) CE4. Time series of $C_6H_{10}O_5$ is shown as an example. CDR, INT, and CF samples are shaded in blue, purple, and gray, respectively. Gaps in the time series represent EESI-ToF-MS background measurement periods, which are not shown.**



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Figure S2. HYSPLIT 3-day backward trajectory analysis of air masses arriving at the Shanghuang site in (a) CE1, (b) CE2, (c) CE3, and (d) CE4. Trajectories are based on cluster analysis for CE1, CE2 and CE4. For CE3, due to its short duration, only 7-hour trajectories are shown with time in UTC.



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Figure S3. Detailed relative contribution of OA. The average carbon number distribution of differences between CDR and CF are colored by oxygen number of CE2 (a), CE3 (b); and nitrogen number of CE2 (c), CE3 (d). Positive value stands for significant molecular characteristics of CDR, and negative value stands for that of CF. Fractions of compounds are normalized to sum of signals of all organics in CDR and CF, respectively.

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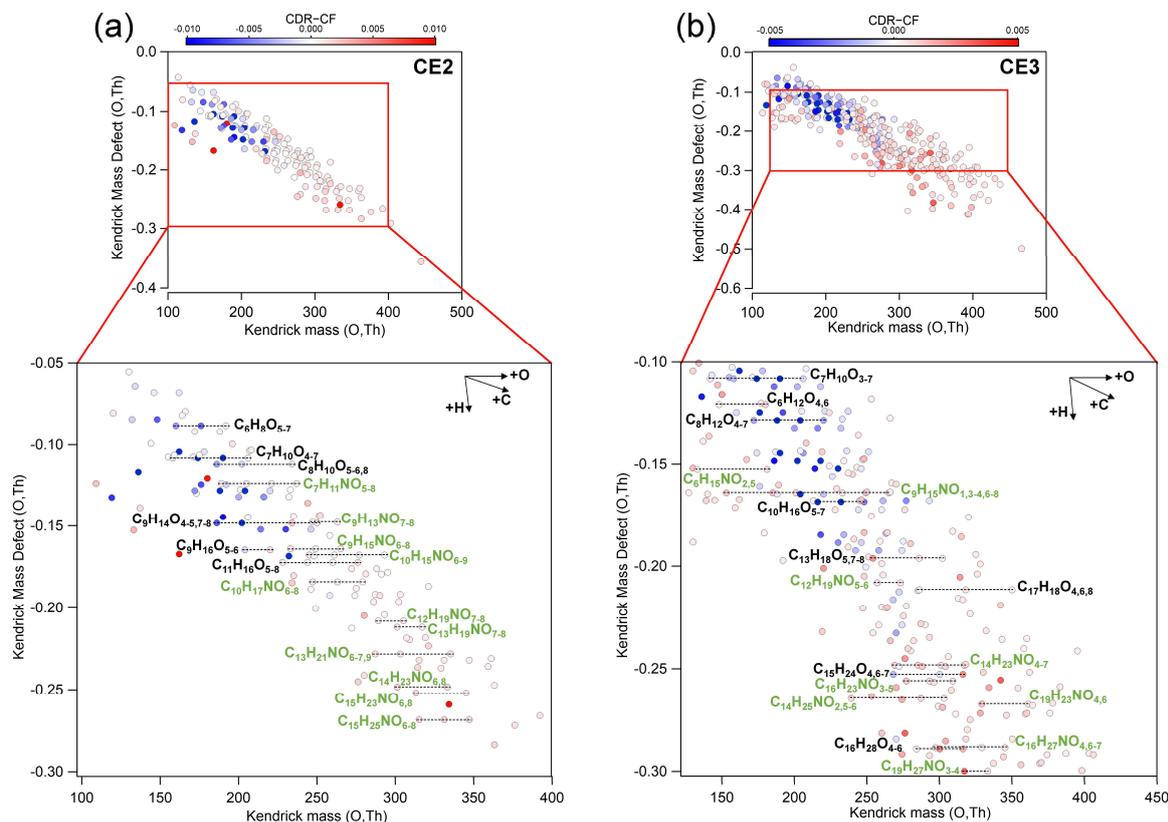
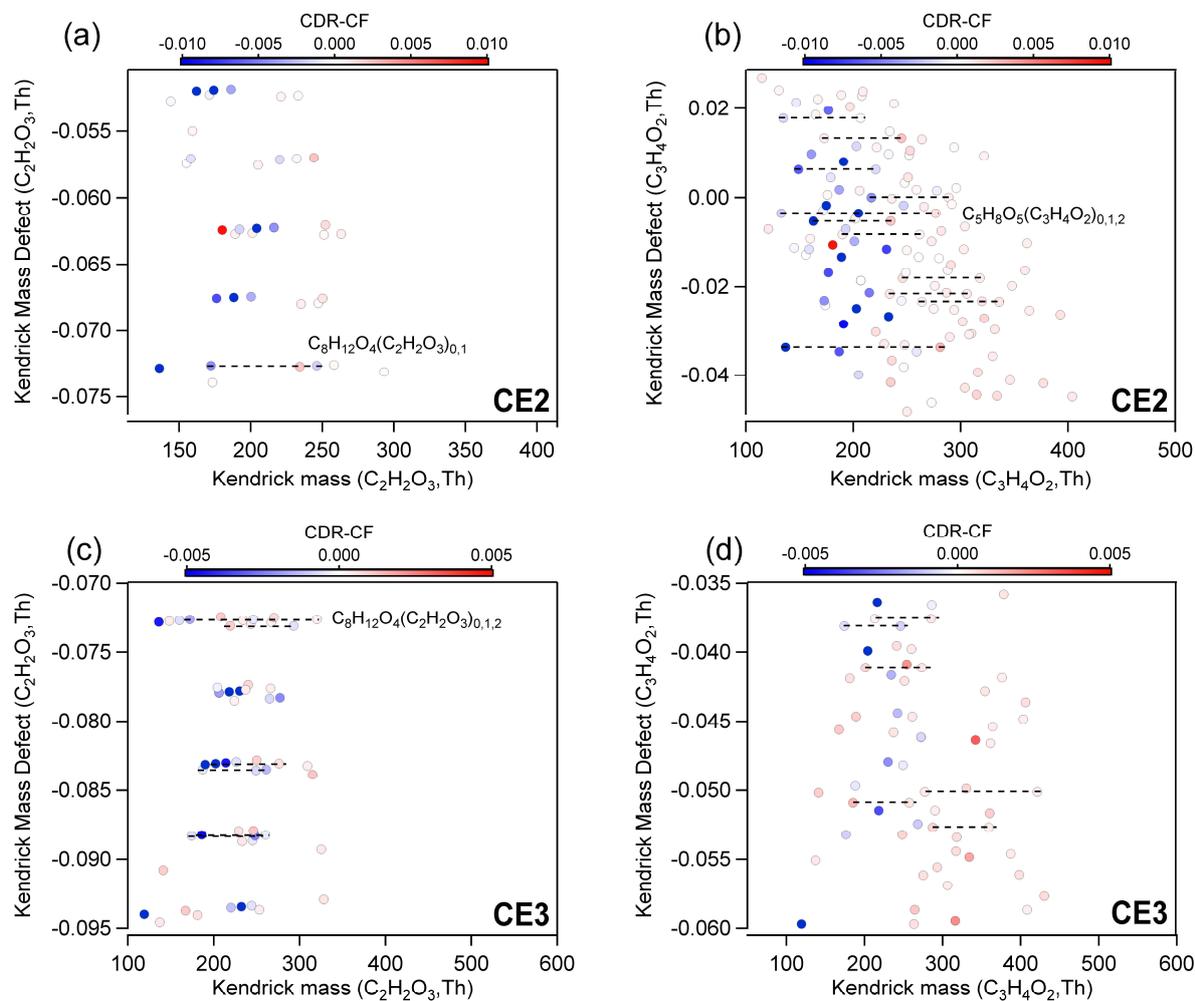


Figure S4. Kendrick mass defect plots based on O of compounds in (a) CE2 and (b) CE3. Data points are color-coded by differences in fraction of compounds between CDR and CF. Fraction of compounds are normalized to the sum of signals of all organics in CDR and CF, respectively. Note that, for conciseness, 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.

40



45 **Figure S5.** Kendrick mass defect plots based on (a) $C_2H_2O_3$ and (b) $C_3H_4O_2$ of compounds in CE2; and (c) $C_2H_2O_3$ and (d) $C_3H_4O_2$ of compounds in CE3. Data points are color-coded by differences in fraction of compounds between CDR and CF. Fraction of compounds are normalized to the sum of signals of all organics in CDR 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.

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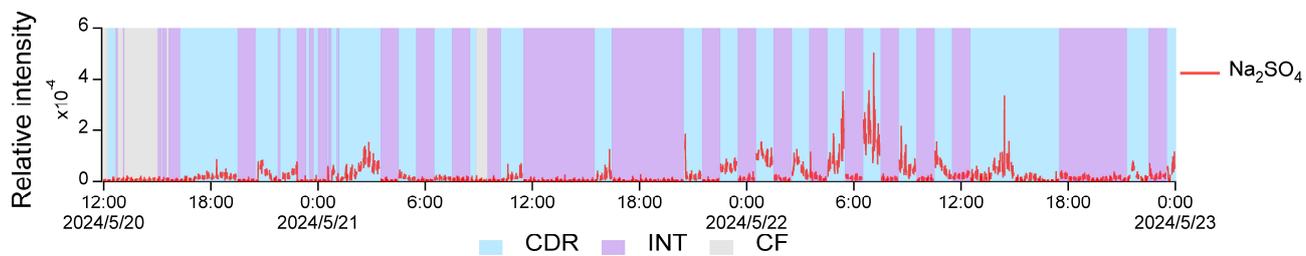


Figure S6. Typical time series of sulfate (Na_2SO_4) in a cloud episode. CDR, INT, and CF are shaded as blue, purple, and gray, respectively. Gaps in the time series represent EESI-ToF-MS background measurement periods, which are not shown.

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Table S1: General characteristics of CE1–CE4, including PM_{2.5} and CO concentration, meteorological parameters (RH and T), origin of air mass, and duration time of each whole cloud episode, respectively. Measured values are shown as mean ± standard deviation.

	CE1	CE2	CE3	CE4
PM _{2.5} (μg m ⁻³)	4.3±3.0	5.3±6.7	13.5±1.5	3.8±3.6
CO (ppm)	0.29±0.03	0.26±0.06	0.17±0.002	0.19±0.03
RH (%)	94.7±12.0	97.7±6.7	90.8±5.9	95.0±9.1
T (°C)	15.1±1.7	15.7±3.0	18.0±0.6	17.3±4.0
Origin of air mass	Southwest	Southwest then turns to north	Southeast	Southwest then turns to northwest
Duration time (hour)	14	22.5	1.5	34

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

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

Table S3: Compounds with larger fraction in CDR than CF in CE1, CE2, CE3, and CE4.

	Formula				
CE1	C4H4NO4	C8H13NO7	C10H17NO6	C15H23NO8	
	C4H6O5	C8H13NO8	C10H17NO7	C15H25NO8	
	C5H13NO2	C8H14O6	C11H16O6	C17H13NO4	
	C5H8N2O3	C9H12O6	C11H17NO7	C18H19NO7	
	C5H12O4	C9H12O7	C11H19NO8	C18H23NO7	
	C6H4O5	C9H13NO7	C11H23NO10	C18H54O9Si9	
	C6H10O6	C9H14O7	C12H19NO7	C19H25NO7	
	C7H8O5	C9H15NO7	C12H19NO8	C19H23NO6	
	C7H8O6	C9H15NO8	C13H15NO6	C21H31NO6	
	C7H10O7	C10H13NO8	C13H19NO7	C23H17NO	
	C7H11NO7	C10H14O7	C15H25NO6	C23H21NO4	
	C7H11NO8	C10H15NO7	C15H25NO7	C25H32O7	
	C8H6O4	C10H15NO8	C15H15O6	C27H20O3	
	C8H10O6	C10H15NO9	C15H15NO7		
	C8H11NO7	C10H16O7	C15H23NO6		
	CE2	C3H2N2O3	C9H8O5	C11H18O6	C14H23NO6
C4H4NO4		C9H12O6	C11H19NO8	C14H23NO8	
C4H8O4		C9H13NO7	C11H20O8	C15H19NO6	
C6H8O7		C9H13NO8	C11H23NO10	C15H25NO6	
C6H9NO7		C9H14O7	C12H12O6	C15H25NO7	
C6H12O6		C9H14O8	C12H16O7	C15H15O6	
C6H15NO2		C9H15NO6	C12H16O8	C15H15NO7	
C7H8O5		C9H15NO7	C12H17NO7	C15H17N3O5	
C7H8O6		C9H15NO8	C12H17NO8	C15H21NO9	
C7H11N		C9H16O6	C12H18O7	C15H22O7	
C7H11NO5		C9H17NO6	C12H18O8	C15H23NO6	
C7H11NO6		C10H8O5	C12H19NO7	C15H23NO8	
C7H11NO7		C10H12O7	C12H19NO8	C15H25NO8	
C7H11NO8		C10H15NO6	C12H24O7	C16H19NO6	
C7H11N3		C10H15NO7	C13H19NO8	C16H48O8Si8	
C7H13N2SO		C10H15NO9	C13H15NO6	C17H13NO4	
C8H6O4		C10H14N2	C13H18O7	C17H22N2O5	
C8H10O8		C10H17NO6	C13H18O8	C18H19NO7	
C8H11NO5		C10H17NO7	C13H19NO7	C18H21NO7	
C8H11NO7		C10H17NO8	C13H20O6	C18H54O9Si9	
C8H14O6		C10H18O6	C13H21NO6	C19H17NO3	
C8H13NO7		C10H19NO6	C13H21NO7	C19H25NO6	
C8H13NO8		C11H14O7	C13H21NO9	C20H24O7	
C8H14O6		C11H16O5	C13H24O6	C21H25NO7	
C9H7N2O		C11H16O7	C14H21NO7	C25H32O7	
C9H7N2O2		C11H16O8	C14H21NO8	C27H20O3	
C9H8SO		C11H17NO7	C14H21NO9		
CE3		C4H7NO3	C12H10O7	C15H22O5	C19H18O6
		C4H7N3O3	C12H13NO6	C15H22O4	C19H19NO2
		C4H8O4	C12H14O7	C15H24O7	C19H20O7
	C4H9N3O3	C12H15NO5	C15H26O6	C19H22O5	
	C5H10O4	C12H16O10	C15H26O6	C19H22O7	
	C5H12O4	C12H19NO5	C15H28O6	C19H23NO4	
	C5H10O4	C12H19NO6	C15H32O7	C19H23NO6	
	C6H4SO3	C12H21NO6	C16H23NO5	C19H24O7	

	Formula			
	C6H10O3	C12H24O7	C16H25NO5	C19H26O4
	C6H11NO5	C12H36O6Si6	C16H27NO4	C19H26O6
	C6H12O4	C13H12O5	C16H27NO6	C19H28O6
	C6H12O6	C13H14O5	C16H27NO7	C19H29O8
	C6H14N2	C13H14O6	C16H29NO5	C19H31NO6
	C6H14N2O	C13H15N3O7	C16H33NO6	C19H38O5
	C6H15NO2	C13H18O7	C16H15NO	C19H39NO4
	C6H15NO5	C13H17NO4	C16H19NO6	C20H29NO5
	C6H15N3O	C13H18O5	C16H22O4	C20H12N2
	C6H17N3O3	C13H18O8	C16H22O9	C20H14N2O2
	C7H10O3	C13H20O6	C16H23NO3	C20H22O5
	C7H11N3	C13H19NO6	C16H23NO4	C20H24O4
	C7H12O8	C13H22O7	C16H24O5	C20H24O6
	C7H15N2	C13H21NO6	C16H25NO8	C20H24O7
	C7H18N2	C13H22O5	C16H26O8	C20H30O10
	C8H15NO4	C13H24O6	C16H28O4	C20H32O9
	C8H4O3	C14H15NO7	C16H28O5	C20H35NO7
	C8H5NO4	C14H23NO7	C16H28O6	C20H36O5
	C8H7NO	C14H7NO4	C16H32O10	C21H21NO6
	C8H13O2	C14H14O4	C17H33NO4	C21H25NO7
	C8H13NO6	C14H16O4	C17H33NO5	C21H22O5
	C8H15N2	C14H16O5	C17H37N5	C21H23NO2
	C9H7NO2	C14H16N2O3	C17H11N	C21H25NO6
	C9H10O2	C14H19NO7	C17H18O6	C21H26O6
	C9H10O2	C14H20O8	C17H18O8	C21H26O8
	C9H13NO2	C14H21NO	C17H22O5	C21H27NO8
	C9H14O7	C14H22O4	C17H22N2O	C21H28O8
	C9H15NO	C14H23NO4	C17H22N2O5	C21H34O2
	C9H15NO3	C14H23NO5	C17H24O5	C21H38O5
	C9H15NO4	C14H23NO6	C17H26N2O	C22H33NO4
	C9H15NO6	C14H24O6	C17H27NO3	C22H26O5
	C9H16O	C14H24O7	C17H28O7	C22H34O7
	C9H19S	C14H25NO2	C17H29NO	C22H38O6
	C10H12O7	C14H25NO5	C17H29NO7	C23H39NO4
	C10H12N2O4	C14H25NO6	C18H25NO4	C23H22O5
	C10H13NO6	C14H26O5	C18H27NO5	C23H26O6
	C10H15NO4	C14H28O5	C18H29NO6	C24H29NO4
	C10H14N2	C14H28O4	C18H16O6	C24H28O5
	C10H20O5	C14H31N3O10	C18H20O6	C24H50O8
	C11H12O4	C15H21NO8	C18H22O5	C25H17NO4
	C11H16O5	C15H25NO5	C18H23N	C26H29O6
	C11H16O8	C15H12O3	C18H27NO7	C26H29NO
	C11H17NO6	C15H11NO2	C18H28O7	C27H38O2
	C11H19NO6	C15H13N3O5	C18H35NO5	C27H50O4
	C11H20O6	C15H15NO2	C18H35N3O8	C28H24O2
	C11H20N2O5	C15H18O8	C18H54O9Si9	
	C11H21NO5	C15H19NO4	C19H27NO3	
	C11H21NO6	C15H20O4	C19H27NO4	
CE4	C3H2N2O3	C9H8SO	C11H16O5	C14H23NO8
	C4H4NO4	C9H8O5	C11H16O6	C15H25NO6
	C4H6O5	C9H10O6	C11H16O7	C15H25NO7

	Formula			
	C4H8O4	C9H12O6	C11H17NO7	C15H12O3
	C5H13NO2	C9H12O4	C11H18O6	C15H15O6
	C5H6O5	C9H12O7	C11H23NO10	C15H20O5
	C5H6O6	C9H13NO7	C12H16O7	C15H22O7
	C5H8N2O3	C9H13NO8	C12H16O8	C15H23NO6
	C5H10O4	C9H14O8	C12H17NO7	C15H23NO8
	C5H12O4	C9H16O6	C12H18O6	C15H25NO8
	C6H10N2O7	C10H8O5	C12H18O7	C17H13NO4
	C7H8O6	C10H12N2O4	C12H18O8	C17H19NO7
	C7H10O7	C10H13NO8	C12H19NO7	C18H19NO7
	C7H11NO6	C10H15NO6	C12H19NO8	C18H21NO7
	C7H11NO8	C10H15NO7	C12H24O7	C18H23NO7
	C7H12O6	C10H15NO8	C13H12O6	C19H25NO7
	C8H12O4	C10H15NO9	C13H18O7	C19H23NO6
	C8H14O5	C10H17NO6	C13H19NO7	C20H31NO8
	C8H14O6	C10H17NO7	C13H20O6	C20H33NO8
	C8H13NO7	C10H18O5	C14H15NO7	C22H25NO6
	C8H13NO8	C10H20O10	C14H21NO8	C25H32O7
	C8H14O6	C11H9NO2	C14H16N2O3	
	C9H7N2O2	C11H14O7	C14H20O6	

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Note: Duplicated molecular formulas originate from different ion adducts from the ionization of EESI-ToF-MS.