

Supplementary Information

Cloud processing and weeklong ageing affect biomass burning aerosol properties over the south-eastern Atlantic

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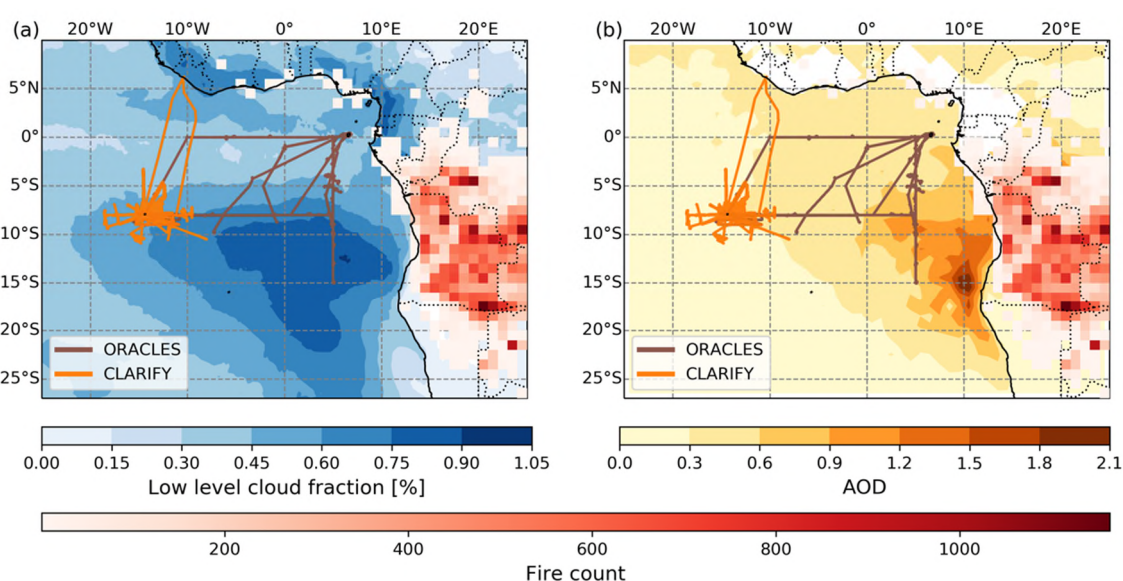
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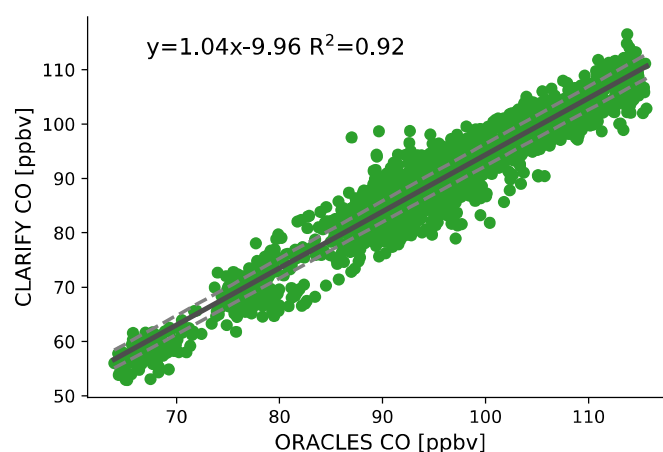


Supplementary figure 1. Accumulated MODIS detected wildfire, CLARIFY and ORACLES (2017) flight track with the mean (a) low-level cloud fraction, and (b) aerosol optical depth during August and September 2017. The wildfire data is from MODIS 6 Collection Active Fire Detections (MCD14ML), distributed from NASA FIRMS [<https://earthdata.nasa.gov/firms>, doi: 10.5067/FIRMS/MODIS/MCD14ML]. Note only fire points with confidence above 80 % are used. The low-level cloud fraction is from ERA5, which is the fifth generation of ECMWF reanalysis data. The AOD data is from MODIS Terra and Aqua 1-degree product at collection 6.

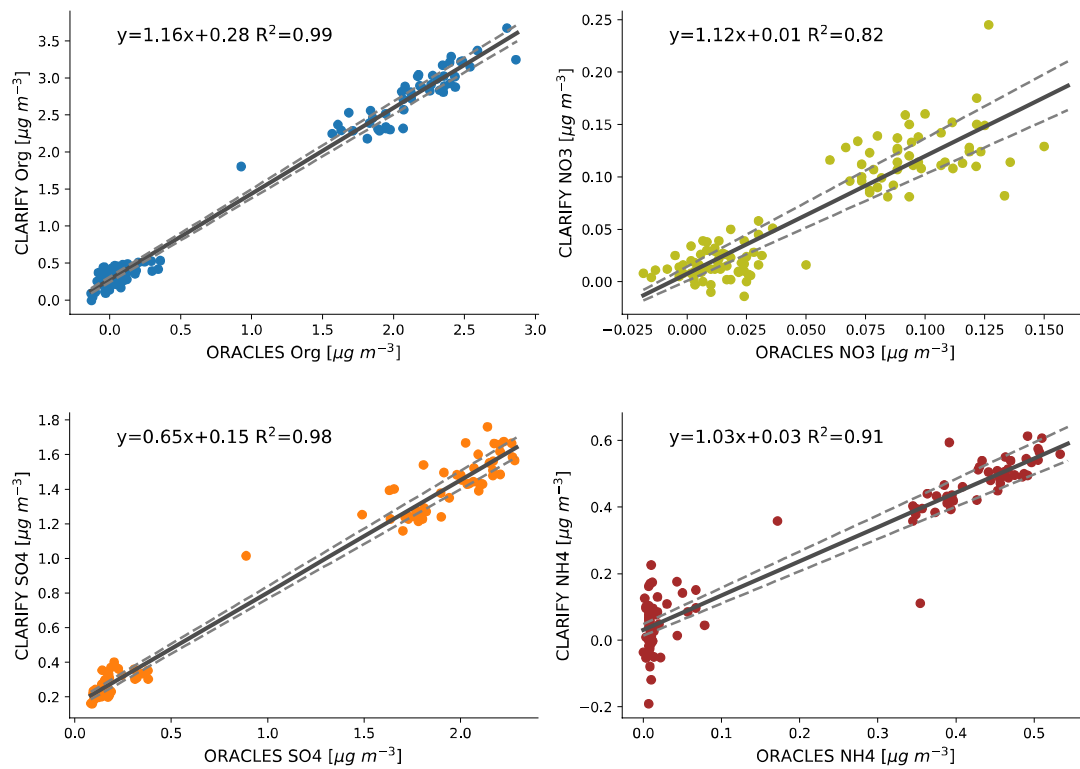
Supplementary Notes 1

CLARIFY and ORACLES intercomparison

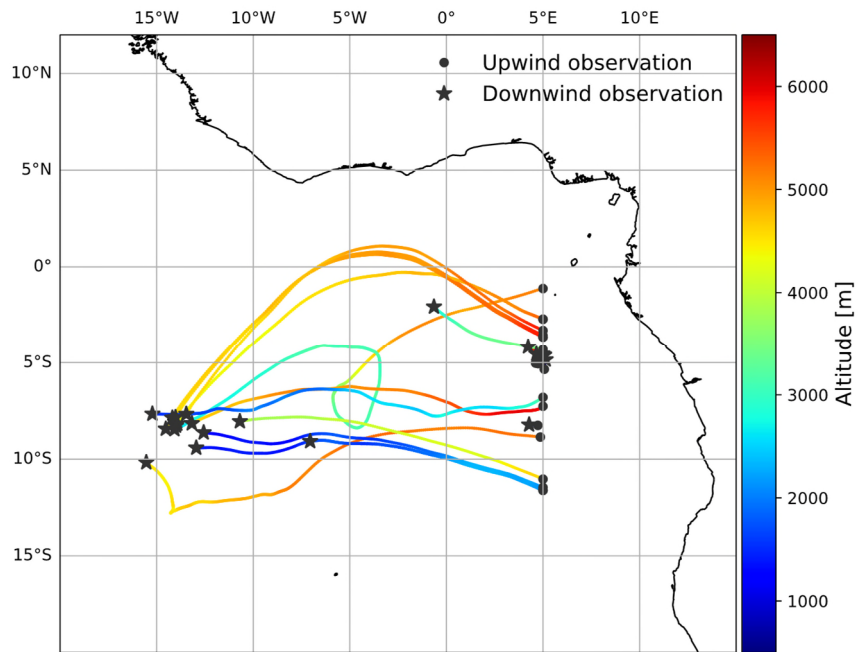
In 20170818, both CLARIFY and ORACLES flew to the same area. However, the air on that day is very clean. Due to the noise, only a few aerosol data are available for comparison. The CO concentrations from the two aircraft are in good agreement (Supplementary Figure 1). Supplementary figure 2 shows the comparison of aerosol chemical compositions measured by AMS. The BC concentration, scattering and absorption are compared by Barrett et al. (2022)¹.



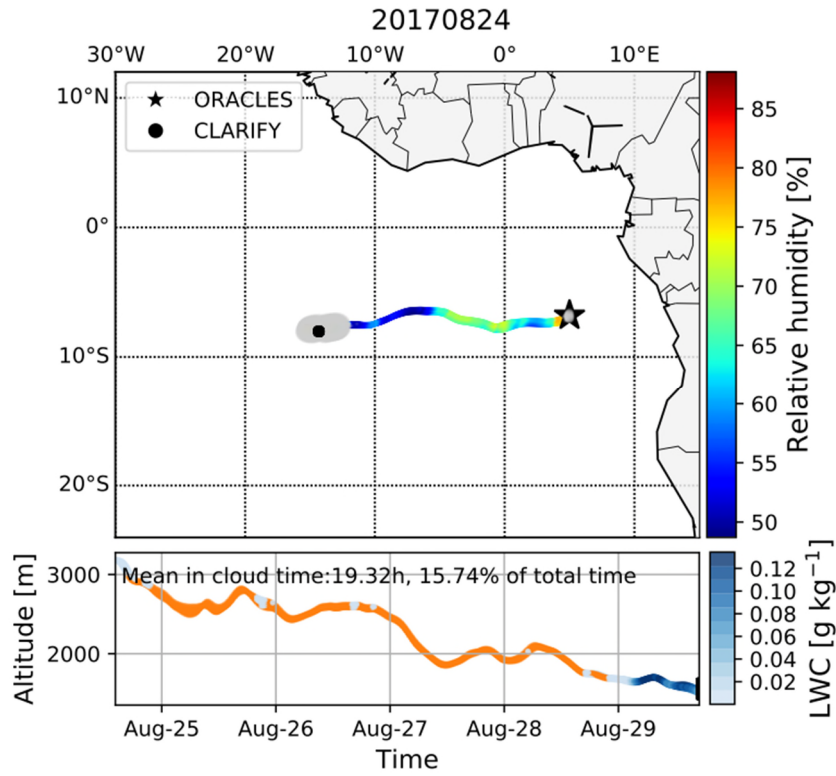
Supplementary figure 2. CO concentrations measured from CLARIFY and ORACLES during the inter-comparison flights.



Supplementary figure 3. AMS data measured from CLARIFY and ORACLES during the inter-comparison flights. Note each data point represents the observation at a 1-second resolution.



Supplementary figure 4. All linked cases. The dots are upwind observations made by ORACLES, while the stars are downwind measurements by ORACLES and CLARIFY. The detailed cases linked with ORACLES and CLARIFY can be found in table S1. The lines connecting upwind and downwind observations are air mass trajectories, and the colour indicates the altitude.

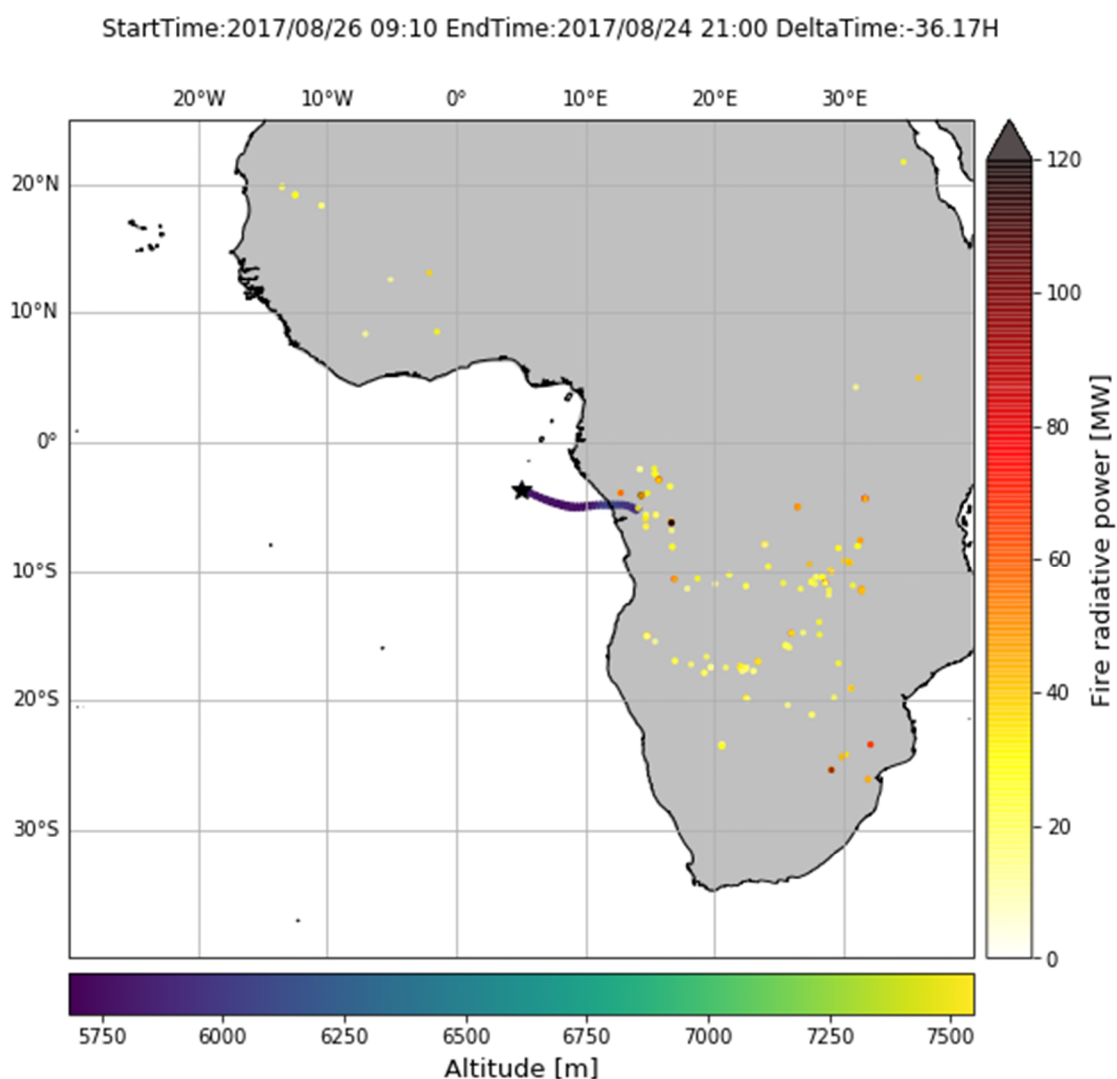


Supplementary figure 5. A linked case of ORACLES and CLARIFY measurements. The star marker represents the location of the ORACLES aircraft, and the dot represents the CLARIFY aircraft. The lines linking ORACLES and CLARIFY are the forward trajectories, and the grey colour shade on the trajectory is the marker for clouds. The lower panel shows the altitude of the trajectories and the cloud liquid water content along the trajectory.

Supplementary Notes 2

Initial age of BB aerosols from ORACLES

We use backward trajectories and fire radiative power (FRP) data to estimate the age of the BB aerosols measured by upwind ORACLES aircraft. The FRP, measured by spinning enhanced visible and infrared imager (SEVIRI) from the geostationary satellite Meteosat-8, is used to estimate the location of the fire². For the linked cases, a backward trajectory is launched from the ORACLES coordinate, using the same ERA5 reanalysis data. Then the age of the BB aerosol measured by ORACLES is estimated when the trajectory intercept with the FRP points. An example of using the backward trajectory to intercept wildfire location is illustrated in Supplementary figure. 6. Here we only select linked cases with an initial ageing time below 2 days to avoid uncertainties caused by different initial ageing hours. The initial age of most of the linked cases is around 40 hours, and can be found in Supplementary Table 1.



Supplementary figure 6. Backward trajectory intercept with the fire, detected with Fire radiative power (FRP) data from the spinning enhanced visible and infrared imager (SEVIRI) on-board the Meteosat-8 geostationary satellite.

Supplementary Notes 3

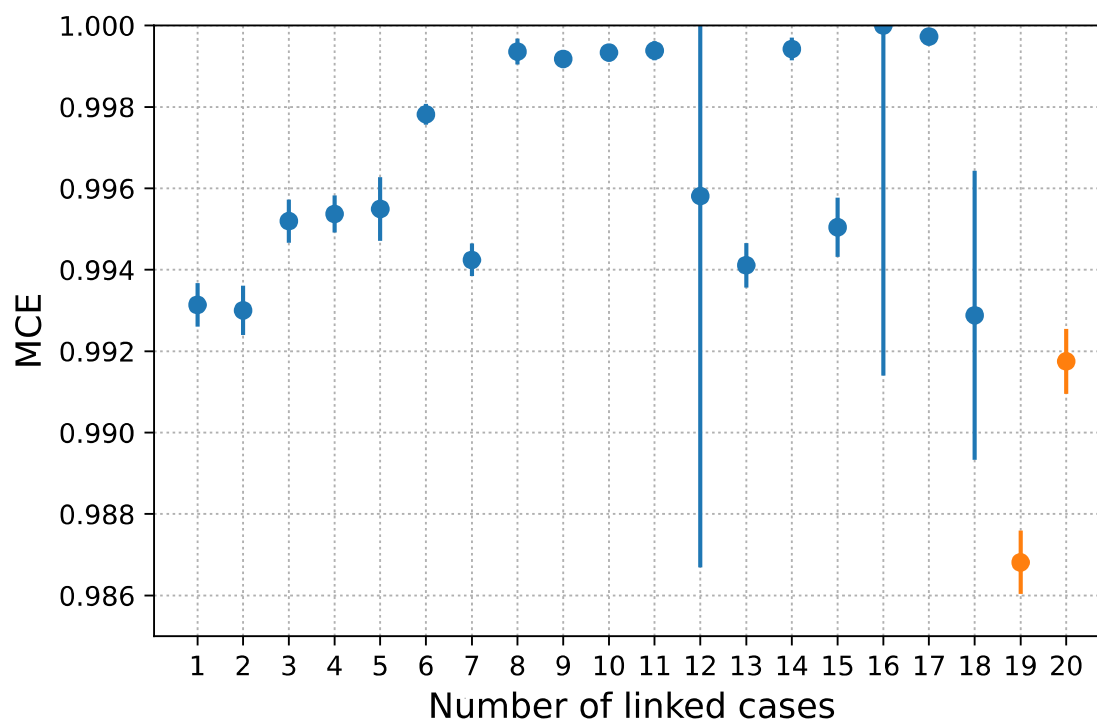
Background CO and CO₂ concentration and Modified Combustion Efficiency (MCE)

The background concentrations of CO and CO₂ are calculated from a combination of methods.

1) Perform linear regression of BC mass with CO and CO₂, as in DeCarlo et al. (2010), by assuming the clean background air has no BC particles. Then the intercept will be the background concentration. 2) Use the 2nd percentile concentrations from the whole campaign by assuming there are no changes in the background concentrations during the 1-month observation.

The calculated background CO₂ are reasonably consistent (~402 ppm), while the CO varied from 65 to 110 ppb, possibly due to measurement uncertainty, and mixing of the plume during transport. To get a realistic background CO, we also compared it with the LASIC observation, which is a ground-based two years campaign on Ascension Island⁸. However, since the LASIC observations are representative of the boundary layer while our upwind observations are generally in the free troposphere, a higher background CO is expected in our results. For this reason, we chose a value of 65.17 ppb (from Method 2) as our background CO. This value is slightly higher, but close to the LASIC result (60 ppb).

Different combustion conditions (flaming, smouldering, or pyrolysis) can result in very different physical and chemical properties of BB emission, which may affect the BB aerosol evolution with ageing^{3,4}. Therefore, we calculated the modified combustion efficiency (MCE) to determine combustion conditions. There are two ways in calculating the MCE 1) $MCE = \Delta CO_2 / (\Delta CO + \Delta CO_2)$, where Δ indicate the difference between observed and background concentration. 2) Perform the linear regression of CO and CO₂, then the MCE can be calculated with the slope of the linear regression: $MCE = 1 / (1 + \Delta CO / \Delta CO_2)$. The advantage of the latter is that it allows the calculation without the background concentration, while the uncertainties in the data can still be passed on to the calculated MCE. Therefore, we compare the MCEs from both methods and find the latter are generally around 1, while both methods yield MCEs higher than 0.98. Thus, the results from the former method are used and illustrated in Supplementary figure 7. This results indicate that the BB aerosols we measured are from fires with flaming combustion, consistent with recent ORACLES and CLARIFY studies⁶.



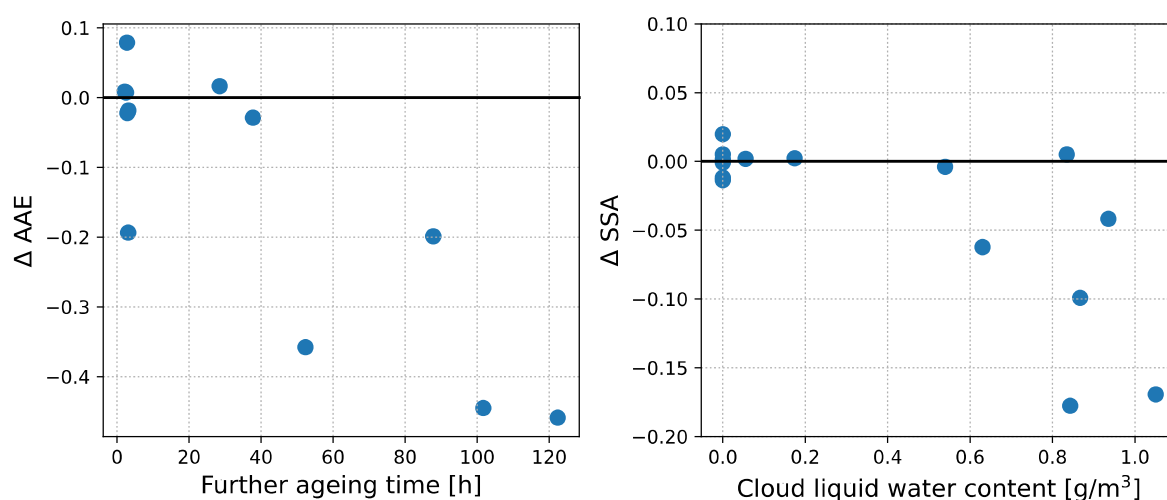
Supplementary figure 7. Calculated MCE for each linked case. The blue dots indicate samples where the mass of OA did not increase with ageing, while the orange dots are two samples where OA increased. The error bars are the standard deviations.

Supplementary table 1. The summary of linked cases. There are 11 ORACLES linked with CLARIFY cases and 12 ORACLES linked with ORACLES cases.

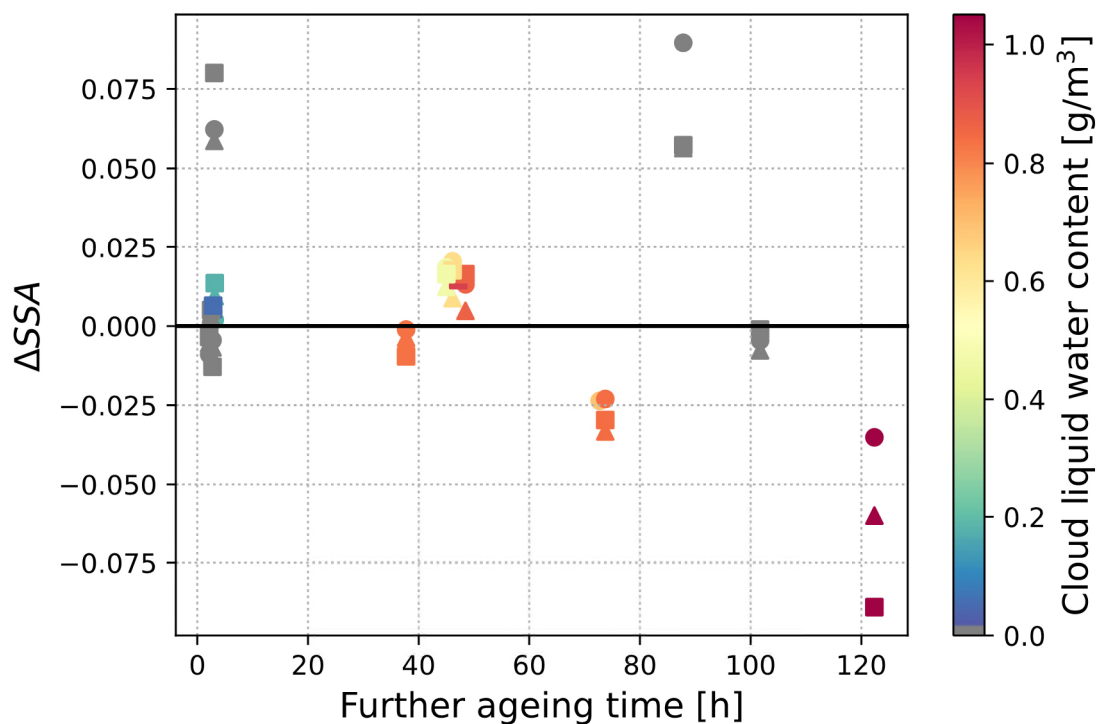
ID	Linked with	Upwind Altitude (m)	Downwind Altitude (m)	Upwind CO (ppbv)	Age of fire at upwind (h)	Transport time (h)	Trajectory distance (km)	Cloud processing *
20170815_1c	CLARIFY	2077	1346	311	46	72.7	2019.7	Y
20170815_2c	CLARIFY	2078	1421	310	44	73.7	2057.8	Y
20170817_1c	CLARIFY	1673	1698	215	48	48.2	909.6	Y
20170817_2c	CLARIFY	1672	1644	211	48	48.5	894.9	Y
20170817_3c	CLARIFY	1670	1669	267	40	45.0	899.9	Y
20170821c	CLARIFY	5791	4572	211	39	87.8	2083.6	N
20170824c	CLARIFY	3314	2247	362	41	122.0	2091.9	Y
20170826_1c	CLARIFY	5322	4499	119	36	77.5	2465.8	N
20170826_2c	CLARIFY	5318	4254	121	32	70.7	2600.1	N
20170826_3c	CLARIFY	5799	4565	114	35	77.5	2596.2	N
20170830c	CLARIFY	4469	3889	489	39	52.3	1788.8	Y

20170813_2o	ORACLES	1524	1598	294	45	2.5	49.0	N
20170815_1o	ORACLES	2078	1683	387	47	47.7	1377.5	Y
20170826_2o	ORACLES	5798	5759	126	38	3.0	98.1	N
20170826_3o	ORACLES	1124	1102	155	38	3.2	56.0	N
20170826_4o	ORACLES	1071	1066	154	40	2.9	51.1	N
20170826_5o	ORACLES	1020	1245	191	39	2.8	59.6	N
20170826_6o	ORACLES	1624	1582	155	38	35	62.7	N
20170826_7o	ORACLES	1020	943	196	38	3.1	67.3	N
20170828_1o	ORACLES	5815	4492	119	39	124.7	2718.0	N
20170828_2o	ORACLES	5800	4554	132	41	118.4	2565.5	Y
20170830o	ORACLES	3560	2818	333	36	28.5	680.1	N
20170831o	ORACLES	3362	2950	362	34	7.2	108.1	N

*Cloud processing is determined if the air mass passes through clouds during the last 24 hours of transport.



Supplementary figure 8. Mie model simulated variations of (a) AAE with further ageing time and (b) SSA with cloud processing. In order to be able to identify, by using the Mie model, whether the observed changes in the optical properties of the aerosol are mainly driven by the variations in the aerosol number size distribution, we used the average refractive index ($1.62 + 0.04j$) from the ORACLES campaign⁹, assuming that the BC is internally mixed, and fixed this refractive index with the observed aerosol size distribution for linked samples for calculation.



Supplementary figure 9. Evolution of single scattering albedo (SSA) with ageing. The grey colour represents samples without cloud processing, and the colourmap indicates the cloud processing intensity.

Supplementary References:

1. Barrett, P. A. *et al.* Intercomparison of airborne and surface-based measurements during the CLARIFY, ORACLES and LASIC field experiments. *Atmospheric Measurement Techniques Discussions* 1–60 (2022) doi:10.5194/amt-2022-59.
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7. DeCarlo, P. F. *et al.* Investigation of the sources and processing of organic aerosol over the Central Mexican Plateau from aircraft measurements during MILAGRO. *Atmos. Chem. Phys.* **10**, 5257–5280 (2010).
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