Supplemental Material:

Characterization of the airborne aerosol inlet and transport system used during the A-LIFE aircraft field experiment

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S1 A-LIFE In-Cabin Instrumentation

Table S1 lists aerosol instruments that were installed in the aircraft cabin of the Falcon and were connected to the isokinetic inlet during the A-LIFE mission. The instrument setup including the flows in the different sampling line parts can be seen in Figure S1. Two experimental instruments which drew together 2.85 1 min⁻¹ are not included in Table S1 and Figure S1.

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In total, the A-LIFE instrumentation drew a volumetric flow of a minimum of 17.87 l min⁻¹ which could increase to a maximum of 22.83 l min⁻¹. The value of the total flow varies because two impactor devices were only turned on during selected measurement periods (typically six times for 5-10 minutes per flight) which increased the total flow by 0.96 l min⁻¹ (Kandler et al., 2007) during these periods. Furthermore, the so-called constant pressure inlet

25 (CPI) system of the DMT Cloud Condensation Nuclei Counter (CCNC) caused a varying flow depending on altitude. The inlet system of the CCNC was used to ensure measurements at a fixed pressure of 500 hPa. The CPI system consists of two orifices with different diameters used at different altitudes, and a pump. Depending on the ambient pressure, the pump regulated the flow (between 0 and 4 1 min⁻¹) so that a pressure of 500 hPa was established behind the orifice.

30 **S2** Aerosol Number Size Distribution

For the derivation of the aerosol number size distribution (NSD) for each of the 262 A-LIFE flight sequences, the data of four instruments were used. The instruments and the size ranges, used for the combined NSDs, are summarized in Table S2.

As explained in Section 2.3 in the main manuscript, the refractive index needed for the derivation of the NSD from 35 OPC measurements is inferred from the aerosol composition along the flight track based on a mixture of five main aerosol types determined with the FLEXPART model (Stohl et al., 1998, Seibert and Frank, 2004). The corresponding refractive indices for each of the five aerosol types are based on literature and are summarized in Table S3. Since the size distribution of CAS is measured at ambient relative humidity conditions, but the other instruments contributing to the combined NSD measure at dry conditions, a growth factor is needed to convert the

40 CAS NSDs to dry particle diameters. The growth factors are dependent on the particle composition, and were also derived based on FLEXPART-modelled aerosol particle composition. The corresponding hygroscopicity of the five aerosol types are also included in Table S3.

| Instrument | Manufacturer | Nominal flow [lpm] | Tubing length [m] | Measured quantity |
|-----------------------------|---------------------------------|-----------------------|----------------------|---|
| CPC1 | TSI | 1 | 5.83 | Integral particle number concentration |
| CPC2 | TSI | 1.5 | 2.66 | Integral particle number concentration |
| CPSA1 | Custom-built at DLR | 1 | 2.39 | Integral non-volatile particle number concentration |
| CPSA2 | Custom-built at DLR | 1 | 2.47 | Integral non-volatile particle number concentration |
| CPSA3 | Custom-built at DLR | 1 | 2.22 | Integral particle number concentration |
| SkyOPC | Grimm | 1.2 | 1.52 | Aerosol number size distribution |
| SkyOPCTD | Grimm | 1.2 | 3.49 | Non-volatile aerosol number size distribution |
| Impactor device 1 | Custom-built at TU Darmstadt | 0.48 | 0.92 | Chemical particle composition, shape |
| Impactor device 2 | Custom-built at TU Darmstadt | 0.48 | 1.02 | Chemical particle composition, shape |
| SP2 (+ Bypass) | DMT | 0.12 (+ 2) | 1.59 | Refractory black carbon mass |
| CCNC (+ CPI) | DMT | 1 (+ 0-4) | 2.85 | Number concentration of cloud condensation nuclei at various supersaturations |
| Aurora 4000 Nephelometer | Ecotech | 2 | 2.68 | Scattering coefficient at three wavelengths (450, 525 and 635 nm) |
| TAP | Brechtel | 2 | 3.52 | Absorption coefficient at three wavelengths (467, 528 and 652 nm) |

45 Table S1: Overview of the in-cabin instrumentation which was connected to the isokinetic inlet during A-LIFE.



Figure S1: Flow plan of the in-cabin instrumentation that was connected to the isokinetic inlet. Note, this flow plan shows the default setup for the A-LIFE campaign which was flown almost the entire time. However, for testing purposes, it was also possible to operate the SkyOPCTD without thermodenuder or behind the constant pressure inlet.

 Table S2: Instrumentation used for the derivation of the aerosol number size distribution (NSD) for each of the 262 A-LIFE flight sequences.

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| Instrument Type | Instrument Model | Size Range Used for Combined NSD | Location | Time Resolution |
|----------------------------------|---|--|--|--------------------|
| Condensation Particle Counter | TSI3760a (CPC2) | $D_p > 10 \text{ nm}$ | In-cabin (particles measured at dry conditions) | 1 Hz |
| Optical Particle Counter | Grimm SkyOPC 1.129 (SkyOPC) | $\begin{array}{l} 280 \text{ nm} < D_p < \ 3 \ \mu\text{m} \\ (\text{for in-cabin NSD}) \\ 280 \text{ nm} < D_p < \ 1 \ \mu\text{m} \\ (\text{for out-cabin NSD}) \end{array}$ | In-cabin (particles measured at dry conditions) | 1 Hz |
| Optical Particle Counter | DMT Ultra High Sensitivity Aerosol Spectrometer – Airborne (UHSAS-A) | $125 \text{ nm} < D_p < 400 \text{ nm}$ | Mounted under the aircraft wing (actively- pumped and dried sample flow) | 1 Hz |
| Optical Particle Counter | DMT Cloud and Aerosol Spectrometer (UNIVIE CAS) | 0.9 μm < D _p < 50 μm (for out-cabin NSD only) | Mounted under the aircraft wing (passive flow; particles measured at ambient relative humidity conditions) | 1 Hz |

| Aerosol Type | Refractive Index | Reference | Hygroscopicity Parameter κ | Reference |
|----------------|--|--|-----------------------------------|---------------------------------|
| Black Carbon | n = 1.75 - 1.95 k = 0.63 - 0.79 | Bond and Bergstrom, 2006 | $\kappa = 0$ | - |
| Sulfate | n = 1.50-1.53 k = 0 | Flores et al., 2012; Tang, 1996; Toon et al., 2006 | <i>κ</i> = 0.483 | Good et al., 2010 |
| Organic Matter | n = 1.44-1.61 k = 0-0.03 | Moise et al., 2015 | <i>κ</i> = 0.163 | Petters and Kreideweis, 2007 |
| Dust | size dependent (see Kandler et al., 2011) | Kandler et al., 2011 | $\kappa = 0.03$ | Herich et al., 2009 |
| Sea salt | n = 1.541 $k = 0$ | Eldrige and Palik, 1985 | $\kappa = 1.1$ | Zieger et al., 2017 |

Table S3: Refractive index (at dry relative humidity) and hygroscopicity parameters used for the derivation of the aerosol NSD including corresponding references. Table modified from Dollner (2022).

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S3 Transport Efficiency

In this study, the transport efficiency was calculated with empirical equations from literature. For Figure 6 in the main manuscript, the inlet efficiency was derived with the experimentally determined sampling efficiency (inlet +
transport efficiency) and the calculated transport efficiency. For this, the transport efficiency of the SkyOPC was used. The transport system for the SkyOPC is summarized in Table S4. The volumetric flow, the length as well as the bend angles were used for the calculation of the efficiency of each tubing part. For the first four sampling line pieces the mean of the flow range was used. The inner diameter of all 9 tubing parts is 4.572 mm.

For the losses of coarse mode aerosol particles in the tubing system, two loss mechanisms were considered: losses in tubing bends and sedimentation losses. For all calculations, the aerosol particle itself was assumed to be a mineral dust particle (density $\rho = 2.6$ g cm⁻³ and shape factor $\chi = 1.2$; Hess et al., 1998 and Kaaden et al., 2008).

S3.1 Particle Losses in Bends

For aerosol particle losses in bends of sampling lines, the following equation given by Pui et al., 1987 was used:

$$\eta_{\text{bend}} = \left(1 + \left(\frac{\text{Stk}}{0.171}\right)^{0.452} \frac{\text{Stk}}{0.171} + 2.242}\right)^{-\frac{2}{\pi}\theta}$$
(S1)

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Here, θ is the angle of curvature of the sampling line in degrees and Stk represents the Stokes number. For the calculation of the Stokes number, the following equations were used (S2-S5; Seinfeld and Pandis, 2016):

$$\eta = 1.7188 \cdot 10^{-5} \left[\left(\frac{\mathrm{T}}{273.15} \right)^{1.5} \left(\frac{384.15}{\mathrm{T}+111} \right) \right]$$
(S2)

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$$\lambda = 0.0651 \frac{\eta}{1.8 \cdot 10^{-5}} \frac{1013}{p} \sqrt{\frac{T}{298}}$$
 (S3)

$$C_{\rm C} = 1 + \frac{2\lambda}{D_{\rm p}} \left[1.257 + 0.4 \exp\left(\frac{-1.1 \, D_{\rm p}}{2\lambda}\right) \right] \tag{S4}$$

Stk =
$$\frac{\rho D_p^2 C_c \left(\frac{V TAS}{7.1}\right)}{18 \eta D \chi}$$
 (S5)

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Here, η is the dynamic viscosity of air, λ the mean free path of the ambient air, Cc the Cunningham slip correction factor. p represents the ambient pressure, while T is the temperature inside the aircraft cabin respectively inside the sampling line, which is assumed to be 30°C. D represents the inner diameter of the sampling line.

105 S3.2 Sedimentation Losses

For aerosol particle losses in bends of sampling lines, the following equation given by Thomas (1958) and Fuchs (1964) was used:

$$\eta_{\text{sed}} = 1 - \frac{2}{\pi} \left(2 \epsilon \sqrt{1 - \epsilon^2} - \epsilon^{\frac{1}{3}} \sqrt{1 - \epsilon^2} + \arcsin\sqrt[3]{\epsilon} \right)$$
(S6)

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$$\epsilon = \frac{3 L v_{TS}}{4 D Q} \cdot \cos \theta$$
 (S7)

given by Heyder and Gebhart (1977). Here, θ is the angle of inclination, L the length of the sampling line, Q the volumetric flow, D the inner diameter of the sampling line and v_{TS} the particle terminal settling velocity, which was calculated with the following equation (Seinfeld and Pandis, 2016):

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$$v_{\rm TS} = \frac{\rho \, D_p^{\,2} \, C_c \, g}{18 \, \eta \, \chi} \tag{S8}$$

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| Sampling line | Flow [lpm] | Length [m] | Bend angle [°] |
|---------------|------------|------------|----------------|
| #1 | 6.90-7.86 | 0.14 | 80 |
| #2 | 6.90-7.86 | 0.12 | 0 |
| #3 | 1.20-2.16 | 0.12 | 0 |
| #4 | 1.20-2.16 | 0.12 | 0 |
| #5 | 1.2 | 0.25 | 90 |
| #6 | 1.2 | 0.30 | 0 |
| #7 | 1.2 | 0.25 | 90 |
| #8 | 1.2 | 0.10 | 0 |
| #9 | 1.2 | 0.12 | 0 |

 Table S4: Overview of all sampling line pieces which formed the transport system of the SkyOPC.

S4 Fitted Ambient Pressure and Temperature

As explained in Section 2.4.3 in the main manuscript, we used the Stokes number Stk_{50} of each v_{TAS} value to convert back to a new cut-off diameter $D_{p,50}$. For this, we used fitted values of ambient pressure and temperature for the whole v_{TAS} range from 70 to 220 m s⁻¹. The used sigmoid fits for this approach are displayed in Figure S2

and Figure S3.



Figure S2: Ambient pressure as a function of v_{TAS} . The points show the 1 s data measured by the CMET system of the Falcon during the A-LIFE campaign. The straight line depicts the sigmoid fit which was used for the calculation of the cut-off diameters.



Figure S3: Ambient temperature as a function of v_{TAS} . The points show the 1 s data measured by the CMET system of the Falcon during the A-LIFE campaign. The straight line depicts the sigmoid fit which was used for the calculation of the cut-off diameters.

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