

Supplementary Material

for

Process-oriented analysis of aircraft soot-cirrus interactions constrains the climate impact of aviation

by

Bernd Kärcher*, Fabian Mahrt & Claudia Marcolli

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Comments:

Supplementary Figs.1-3 show results from the soot-PCF model (Sensitivity studies in S.Fig. 2)

Supplementary Figs.4-8 illustrate results from the cloud model (Sensitivity study in S.Fig. 8)

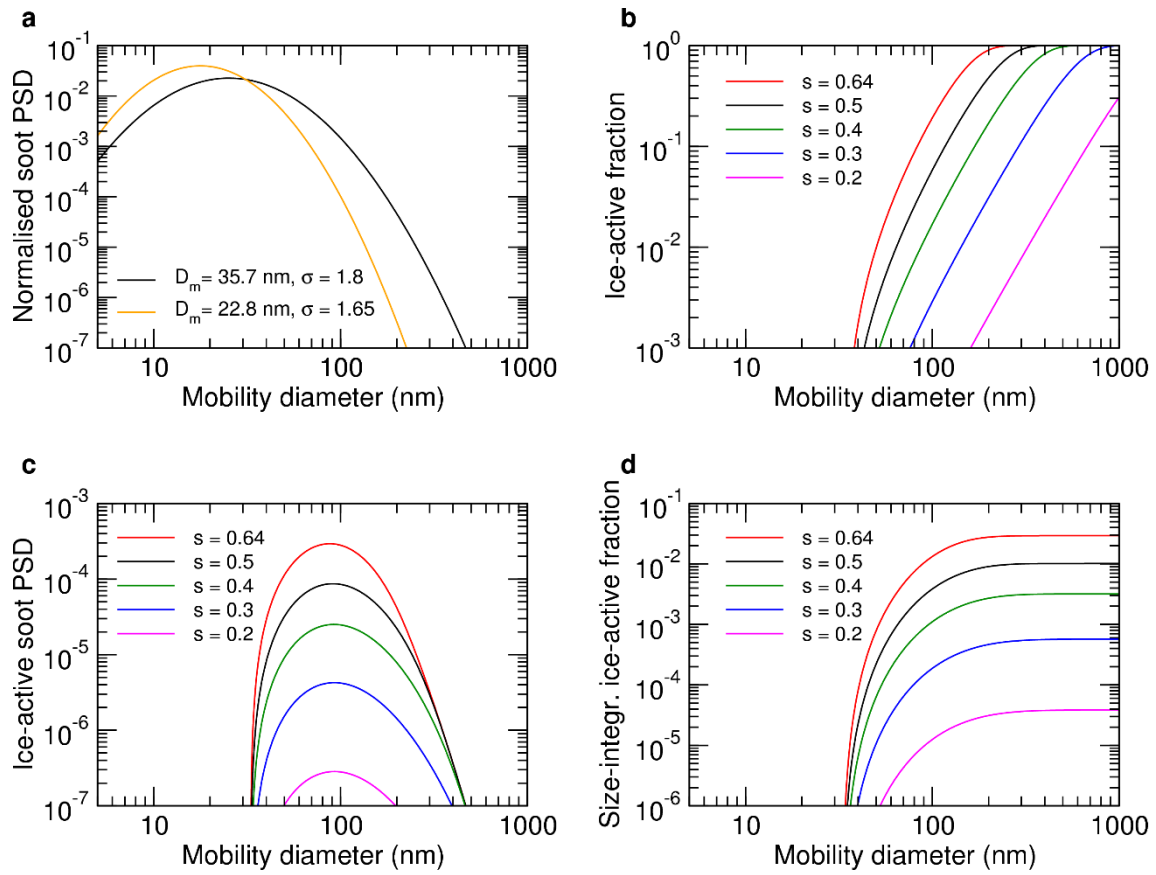
Supplementary References appear in the publication and are added here for convenience

* Corresponding author email: Bernd.Kaercher@dlr.de

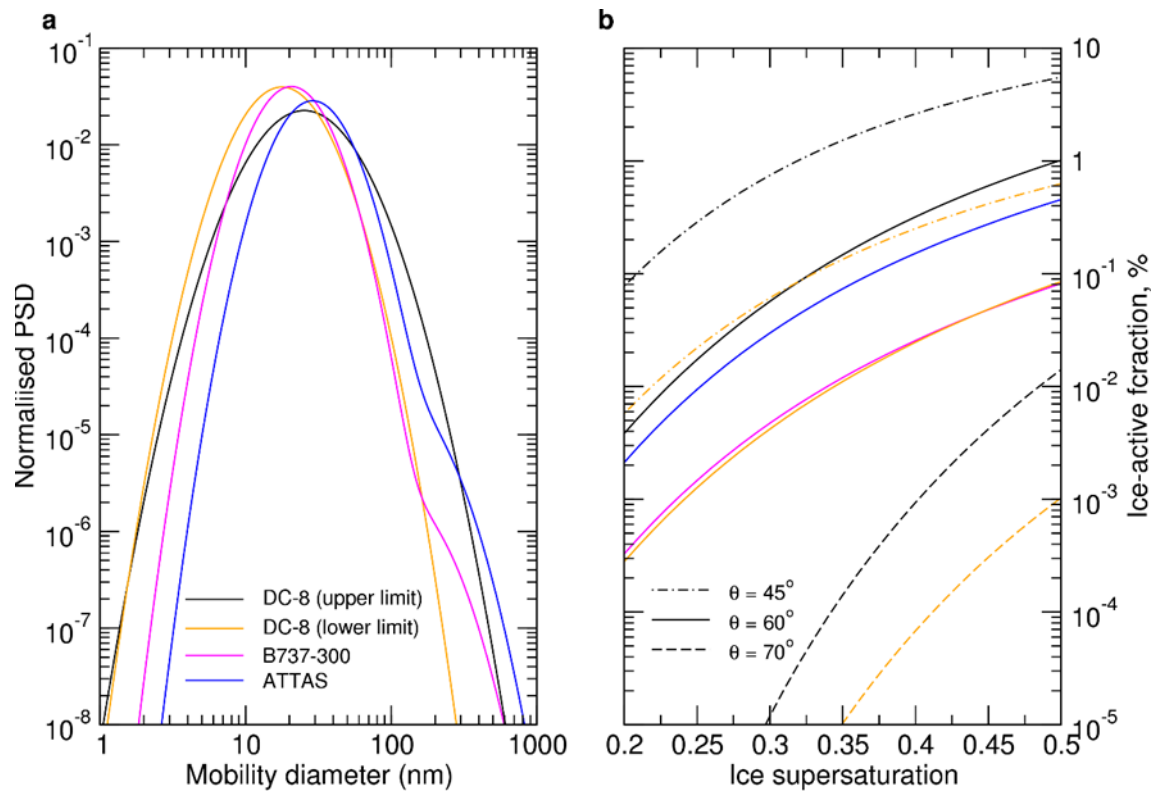
Supplementary Table 1. Parameters used to initialise the 1D-cirrus simulation scenarios

Simulation scenario	Plume age (h)	Soot layer depth (m)	Soot number conc. (L^{-1})	Vertical diffusivity ($\text{m}^2 \text{s}^{-1}$)	Entrainment rate (h^{-1})
Base	—	0	0	0	0
Background	5	400	458	0	0.215
Plume	0.5	300	5448	0.15	2.15

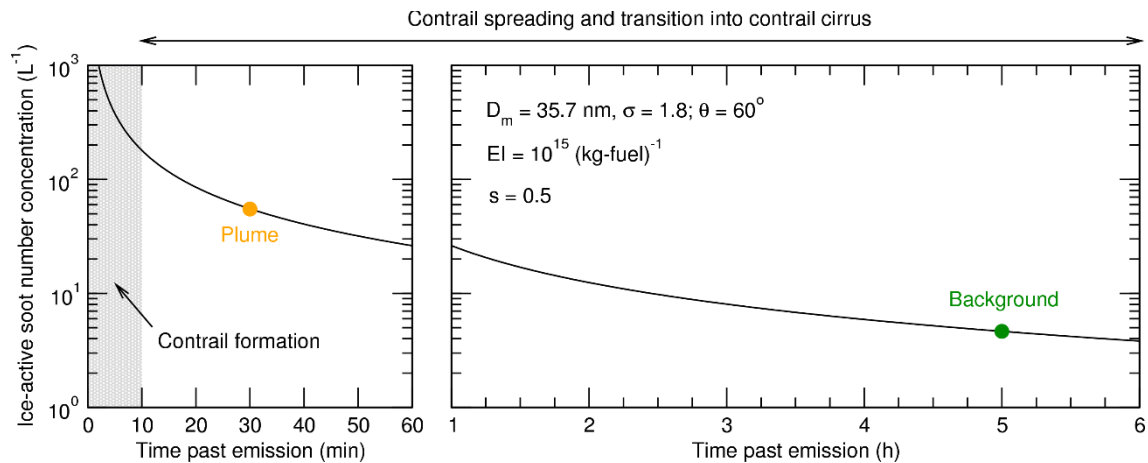
Scenario Base describes unperturbed cirrus formation by homogeneous freezing of supercooled aqueous solution droplets. In the Plume and Background scenarios, aircraft soot particles are also allowed to form and grow cloud ice crystals via PCF. Plume age refers to the time past emission. The instant when ice saturation is first reached across the initial moisture profile at 10 km altitude sets the initial time ($t = 0$) of the numerical simulations. Typical soot particle layer depths are estimated from vertical plume extensions. Only a small fraction of emitted soot particles is ice-active (Supplementary Fig.1). Plume diffusivity is based on large-eddy simulations of aircraft emissions in young exhaust plumes in the stratified upper troposphere; diffusivity is small in background and ambient conditions and is therefore neglected. The initial rate of entrainment of ambient air into the cloud column diminishes linearly with time in the simulation. Soot particles emitted at cruise are log-normally distributed with total number emission index of $10^{15} (\text{kg-fuel})^{-1}$, modal diameter of 35.7 nm and geometric standard deviation of 1.8. The soot-PCF parametrization^{S1} assumes a contact angle at the soot-liquid water interface of 60° . These values are varied in sensitivity studies (Supplementary Fig. 2).



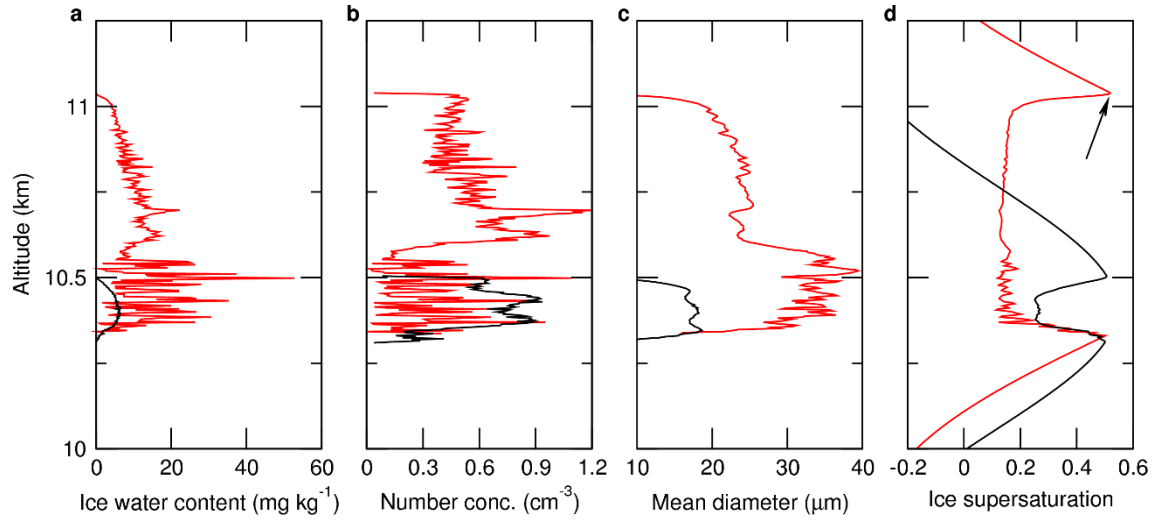
Supplementary Figure 1. Properties of ice-active aircraft soot particles. **a** Normalised PSDs of freshly emitted exhaust soot particles measured at cruise altitudes (9–11 km)^{S2} versus dry mobility diameter, D . Colours represent (black) upper-limit and (orange) lower-limit values for the modal diameter, D_m , and geometric standard deviation, σ . Only 0.9% (black) and 0.1% (orange) of all particles have diameters > 200 nm. **b** Ice-active soot fractions, $AF(D,s)$, predicted by the soot-PCF parametrization at a typical cirrus temperature of 220 K for different values of ice supersaturation, s , up to liquid water saturation (≈ 0.64). AF -values are cumulative in s and based on a soot-water contact angle of 60° . **c** Ice-active soot PSDs, $(AF \cdot PSD)$, reveal significant abundance only for particles with D in the range 50–200 nm. **d** Ice-active soot particle fractions obtained by integrating the ice-active PSDs from panel **c** over D . All curves in panels **c,d** are based on the (black) upper-limit PSD.



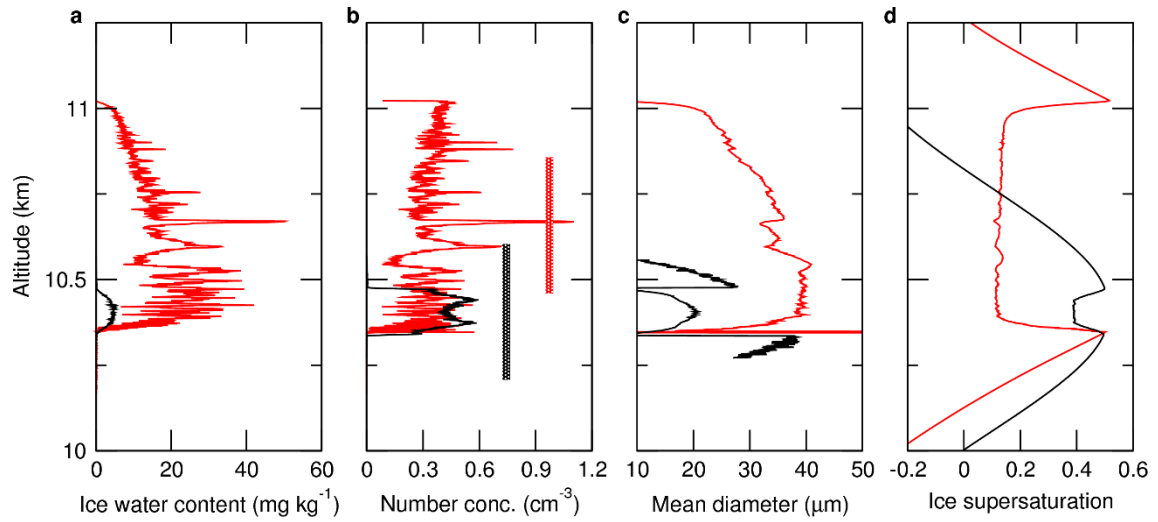
Supplementary Figure 2. Ice activity spectra of aircraft soot particles. **a** Monomodal^{S2} (black, orange) and bimodal^{S3} (blue, magenta) soot PSDs from in situ measurements in young exhaust plumes (without contrails) of different types of aircraft. The monomodal PSDs are repeated from Supplementary Fig. 1a to facilitate comparison. **b** Spectral distributions of ice-active fractions derived from the PSDs. Ice activity is predicted by the soot-PCF parametrization assuming a temperature of 220 K for (solid curves) the default soot-water contact angle, θ , of 60° . Results for (dashed) higher and (dot-dashed) lower θ -values are shown only based on the two monomodal PSDs. The solid black activity spectrum in panel **b** is consistent with the size-integrated results shown in Supplementary Fig. 1d in the limit of large mobility diameter.



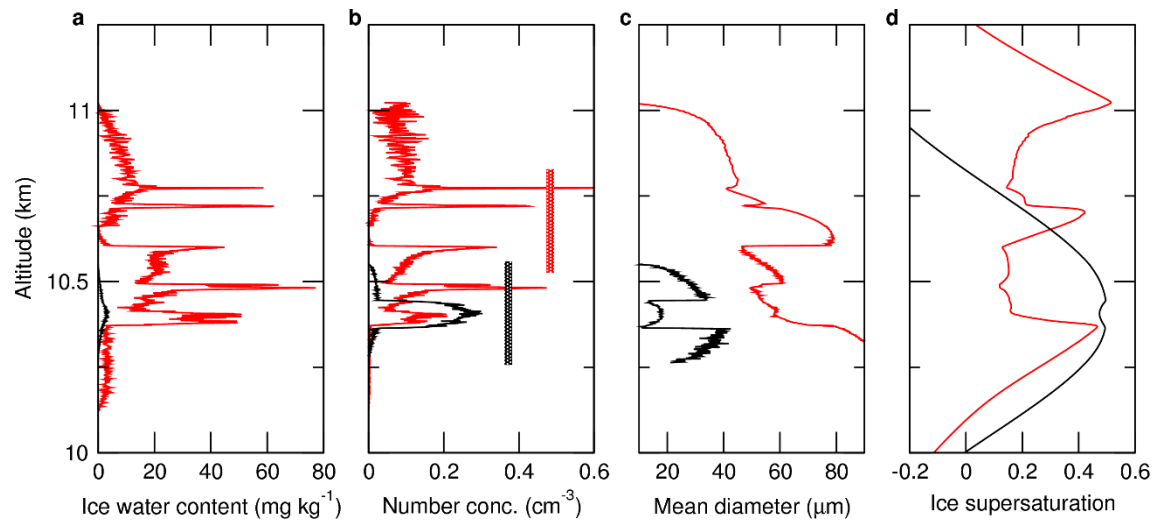
Supplementary Figure 3. Temporal evolution of ice-active aircraft soot particle number concentrations. Number concentrations predicted by the soot-PCF parametrization at the onset of homogeneous aerosol freezing in a diluting aircraft plume as a function of time past emission (plume age), t , in the contrail formation ($t \leq 10 \text{ min}$) and spreading stages. Results are obtained for a soot particle number emission index of $EI = 10^{15} \text{ (kg-fuel)}^{-1}$. Based on a single plume dispersion model^{S4}, soot particles are diluted at cruise altitudes while moving away from the exit plane of an aircraft jet engine. The calculations assume a modal diameter, $D_m = 35.7 \text{ nm}$, and geometric standard deviation, $\sigma = 1.8$, of log-normally distributed soot particles; only the ice-active portion is accounted for, cumulated up to an ice supersaturation of $s = 0.5$ and based on a soot-water contact angle, $\theta = 60^\circ$. Circles mark the analytical estimates, taken at plume ages of $t = 0.5 \text{ h}$ and 5 h , that are associated with the simulation scenarios Plume and Background, respectively.



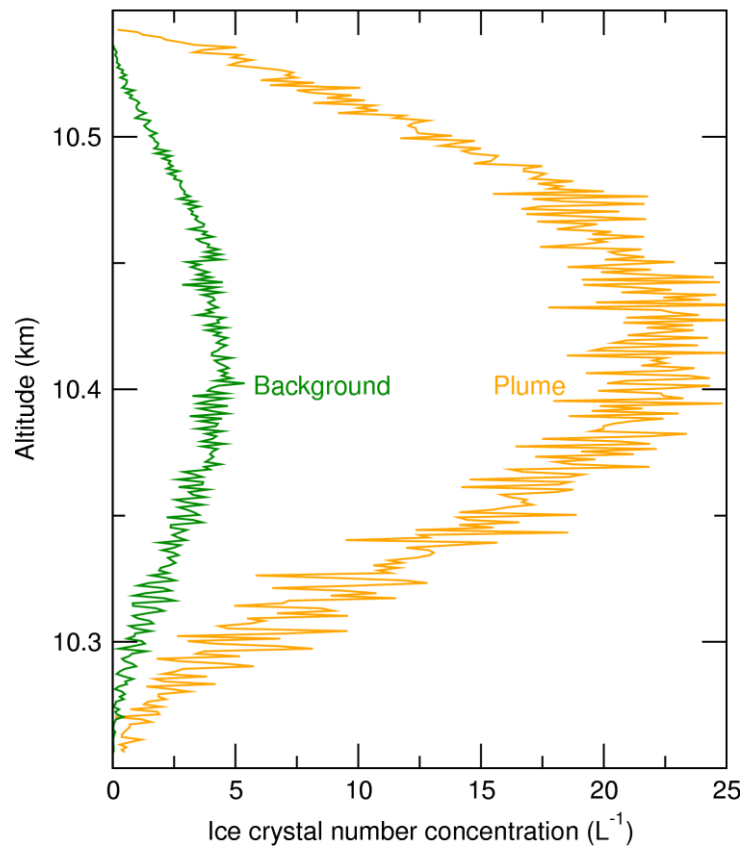
Supplementary Figure 4. Unperturbed cirrus simulation Base. Vertical distributions of **a** ice water content; **b** ice crystal number concentration ($1 \text{ cm}^{-3} = 1,000 \text{ L}^{-1}$); **c** number-weighted mean ice crystal diameter; **d** ice supersaturation. The coloured curves show results for homogeneous aerosol freezing in a constant updraught of 0.15 m s^{-1} , taken at (black) 45 min and (red) 75 min. The lifting distance to reach first homogeneous freezing conditions is approximately 400 m. Further ice nucleation at the cloud top (arrow in panel **d**) and subsequent ice crystal growth and sedimentation produces at 75 min an extended cloud layer between about 10.3 km and 11 km. Narrow spikes in ice water content or ice crystal number concentration are transient features that appear when many ice crystals sediment by chance into the same altitude grid cell (resolution: 1 m). Low values (< 0.01) of the deposition coefficient (not shown) limit ice crystal growth^{SS}; together with sustained cooling, this results in significant ice supersaturation in the cloud core (0.15–0.2).



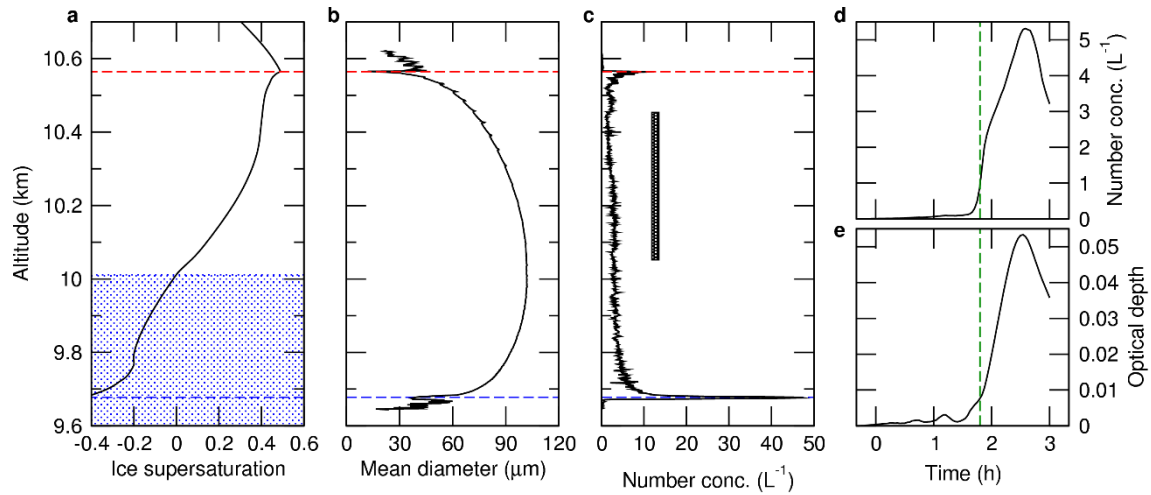
Supplementary Figure 5. Perturbed cirrus simulation Background. Vertical distributions of **a** ice water content; **b** ice crystal number concentration; **c** number-weighted mean ice crystal diameter; **d** ice saturation ratio. Aircraft soot particles form ice via PCF. Colours mark results taken (black) 45 min and (red) 75 min past initialisation. Coloured bars in panel **b** mark the extent and actual position of the soot aerosol layer.



Supplementary Figure 6. Perturbed cirrus simulation Plume. Vertical distributions of **a** ice water content; **b** ice crystal number concentration; **c** number-weighted mean ice crystal diameter; **d** ice saturation ratio. Aircraft soot particles form ice via PCF. Colours mark results taken (black) 45 min and (red) 75 min past initialisation. Coloured bars in panel **b** mark the extent and actual position of the soot aerosol layer.



Supplementary Figure 7. Soot-derived ice crystal number concentrations in perturbed cirrus simulations. Vertical profiles of the number concentrations of ice crystals derived from contrail-processed aircraft soot particles. Results are taken right before homogeneous freezing sets in (including plume dilution since ice saturation) and therefore represent maximum ice activity. Peak values of about 4.5 L^{-1} (22 L^{-1}) are attained in scenario Background (Plume), at maximum ice supersaturation near 10.4 km altitude.



Supplementary Figure 8. Perturbed cirrus simulation Background with low updraught speed. Vertical distributions at 3 h past the time of first soot-induced ice crystal formation via PCF of **a** ice supersaturation; **b** number-weighted mean ice crystal diameter and **c** ice crystal number concentration. Temporal evolution of **d** column-averaged ice crystal number concentration and **e** short-wave cirrus optical depth; while the simulation starts at ice saturation, results are plotted vs time after first soot-induced ice crystal formation. The imposed updraught speed is 0.01 m s^{-1} . The blue-shaded area in panel **a** indicates the ice-subsaturated cloud layer. Dashed lines in panels **a-c** mark the sublimation peak (blue) at the cloud base and homogeneous freezing (red) at the cloud top. The vertical bar in panel **c** marks the extent and actual position of the soot aerosol layer. Dashed lines (green) in panels **d** and **e** mark the time after which homogeneous freezing is the dominant ice production mechanism. Owing to the low updraught speed, soot-induced ice crystal growth starts quenching ice supersaturation already at a slightly lower supersaturation due to a larger sizes of freezing droplets, suppressing the homogeneous freezing burst seen around 10.4 km in Supplementary Fig. 5. After about 2.5 h, sublimation diminishes total ice crystal number concentration and cloud optical depth.

Supplementary References

- S1. Marcolli, C., Mahrt, F. & Kärcher, B. Soot-PCF: Pore condensation and freezing framework for soot aggregates. *Atmos. Chem. Phys. Discuss.*, doi:10.5194/acp-2020-1134 (2020).
- S2. Moore, R. H. et al. Biofuel blending reduces particle emissions from aircraft engines at cruise conditions. *Nature* **543**, doi:10.1038/nature21420 (2017).
- S3. Petzold, A., Döpelheuer, A., Brock, C. A. & Schröder, F. In situ observations and model calculations of black carbon emission by aircraft at cruise altitude. *J. Geophys. Res.* **104**, doi:10.1029/1999JD900460 (1999).
- S4. Kärcher, B., Burkhardt, U., Bier, A., Bock, L. & Ford, I. J. The microphysical pathway to contrail formation. *J. Geophys. Res.* **120**, doi:10.1029/2015JD023491 (2015).
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