

Homogeneous ice nucleation from water vapour suggested by elongated clouds on Mars

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Supplementary discussion S1. Details on quantitative comparison between mesoscale model and observations

Here we give some additional details to the comparison given in the main text. Extended Data Figure 4 and Movie S2 are a support for this comparison. Observed parameters are extracted from Hernández-Bernal et al.²⁵ except otherwise indicated.

The observed cloud width is around 120 km, in our reference homogeneous simulation it is around 50 km (see HOMO_HTJSC). It grows considerably in other simulations in which homogeneous nucleation is favoured, by reducing to surface energy or the vapor pressure of ASW (see for example simulations SIGMA_095 or LOWVAP). When nucleation increases, the radius of the nucleation volume expands, leading to a wider cloud tail.

Observations show that the cloud expansion starts at 5:40 Local Time, with sunrise, and it reaches a length of up to 1800km. In the model the cloud expansion starts between 6:00 (LOWERVAP1) and 7:20 (HOMO_HTJSC).

The observed cloud reaches a length of up to 1800km. The length in the reference homogeneous simulation (HOMO_HTJSC) is around 450 km, and in other simulations it reaches a maximum of 800km (LOWERVAP1). The reasons for this discrepancy are discussed in the main text.

The observed optical depth displays large daily variations, as shown in Methods, ranging from 0.2 to 0.7 at least. In comparison, the reference homogeneous simulation (HOMO_HTJSC) produces a peak optical depth of 0.36 (fig. S4). Other simulations within a realistic range of uncertainties (LOWVAP, SIGMA_095) produce an optical depth of 0.6-0.8.

The size of homogeneously formed ice particles in the AMEC is around 0.25 μm in the reference homogeneous simulation (fig. S4). While no measurements of this parameter are available for the AMEC in particular, Clancy et al.²⁷ reported particle sizes of 0.2-0.5 μm for the Perihelion Cloud Trails, which are, as discussed in the main text, similar in many senses to the AMEC, and therefore this is possibly a good indication of the particle sizes of the AMEC.

Supplementary discussion S2. Effects of hypothetical submicrometer-sized aerosols

In order to simulate cloud formation on a bimodal dust distribution³⁶⁻³⁸ and on dirty ice particles formed on Meteoric Smoke Particles³⁹ (MSPs from now onwards), we introduced a secondary distribution of aerosols able to nucleate heterogeneously in the mesoscale model (see Methods). We also used for this analysis the 0D microphysics model. Using these tools we investigated the possibility that the AMEC is the result of heterogeneous nucleation on submicrometer or nanometer sized dust.

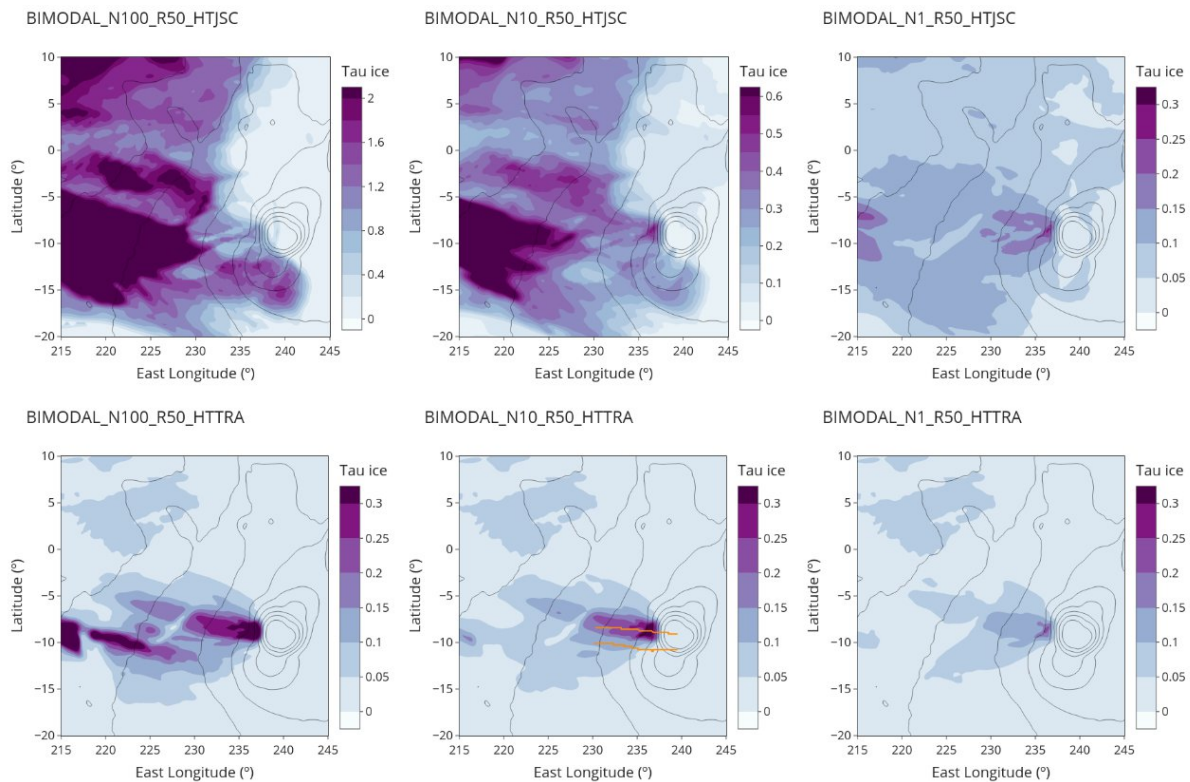


Figure S1. Optical depth obtained in mesoscale simulations when including the finer dust distribution with an effective radius of 50nm. All plots correspond to 8:05 a.m. Local Time. Only heterogeneous nucleation is simulated. Lower-middle panel includes two orange curves representing the trajectories of the Lagrangian cells simulated in the 0D model.

Figure S1 shows cloud optical depth obtained in mesoscale simulations for various scenarios of finer dust distribution (varying number densities, fixed radius of 50nm, as reported by Fedorova et al.³⁶) and contact parameters (see Methods and Extended Data Table 1 for details on each simulation). We see that for the HTJSC contact parameter widespread hazes form without any resemblance to the observed AMEC. When using the HTTRA hazes downwind from Arsia Mons are thicker than elsewhere, but unrealistic hazes remain in these simulations. By contrast, HRSC observations (upper panels in figure S2) do not show thick hazes around the AMEC, and quantitative results by Fernando et al.⁶⁸ (their Fig. 9, row 1, third panel) show clearly the high contrast between the brightness of the AMEC ($\tau \sim 0.7$) and the lack of thick hazes around it ($\tau \sim 0.1$). Our own measurement on another picture (see Methods) suggests $\tau \sim 0.2-0.3$ for the AMEC tail.

We cannot count during this season with existing observational constraints for the actual size distribution and number density of fine dust particles. In order to explore the whole parameter space, we used the 0D microphysics model, which is hundreds of times faster than running the full mesoscale model, to run a large number of simulations using different combinations of parameters (Table S1).

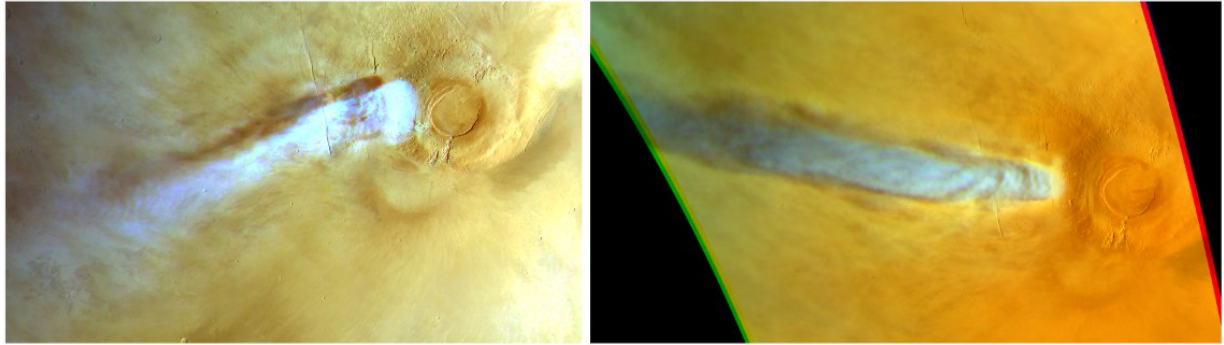


Figure S2. Observations for comparison to simulations. (Left) MEX/HRSC observation G486. (Right) MEX/HRSC observation P802.

Parameter	Tested values	Number of values
Contact parameter	HTTRA, HTJSC, 0.95	3
N1 (cm ⁻³)	0.01, 0.1, 1, 10	4
R1 (μm)	0.5, 1, 1.5, 2	4
N2 (cm ⁻³)	0.1, 1, 10, 100, 1000	5
R2 (nm)	1, 5, 10, 50, 100, 500	6

Table S1. Values considered in our exploration of the space of parameters for the bimodal dust distribution. The total amount of combinations is 1440=3x4x4x5x6. N1, and R1 refer to the number density and radius of the coarse dust mode. N2 and R2 refer to the smaller dust mode. The effective variance of each log-normal distribution is kept constant $v_{eff} = 0.4$.

For each scenario, we ran two different 0D simulations for two different Lagrangian cells extracted from the mesoscale model (see the trajectories of these lagrangian cells in figure S1):

- Tail cell: traverses the core cold pocket, and so it represents cloud formation in the location where the cloud tail is observed. Provides the reached cloud optical depth (τ_{tail}).
- Hazes cell: moves ~100km South from the core cold pocket, and representing the formation of hazes around the AMEC. Provides the reached cloud optical depth (τ_{hazes}).

Then we make dispersion graphs showing, for each scenario, the optical depth of the tail (τ_{tail}) versus the ratio of the optical depth in the tail and outside of it (τ_{tail}/τ_{hazes}). In the observations of Fernando et al.⁶⁸, $\tau_{tail} \sim 0.7$ and $\tau_{tail}/\tau_{hazes} \sim 7$, other other observations are compatible with lower optical depths (see Methods). A potentially realistic reproduction of the AMEC would correspond to a more or less elevated optical depth in the tail ($\tau_{tail} > 0.3$) and a

high contrast between the optical depth of the tail and the hazes ($\tau_{tail}/\tau_{hazes} > 4$). This is a simplified metric, but a success in this metric does not guarantee a success in terms of morphology and dynamics in the mesoscale model, and so this metric is only used for the exploration of the parameter space.

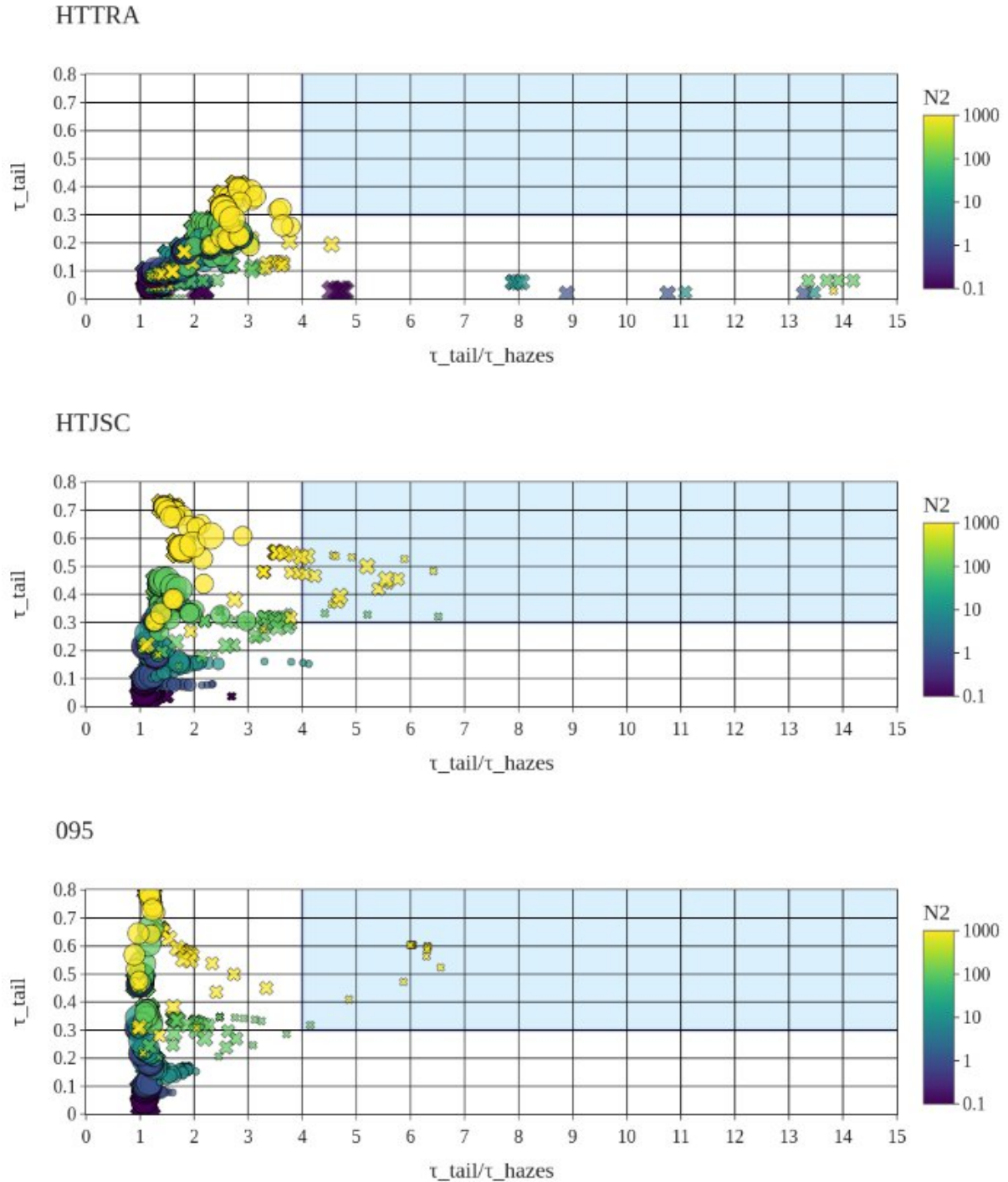


Figure S3. Results from 0D simulations for the parameter space from table S1, including the bimodal distribution and only heterogeneous nucleation. The area corresponding to potentially realistic reproductions of the AMEC ($\tau_{tail} > 0.3; \tau_{tail}/\tau_{hazes} > 4$) is indicated by a light blue background. Unrealistic scenarios ($N_2 \geq 100 \text{ cm}^{-3}$ and $R_2 \leq 10 \text{ nm}$, or $N_1 = 0.01 \text{ cm}^{-3}$. See text) are indicated by crosses. Marker size is proportional to R_2 in each scenario.

Homogeneous nucleation was deactivated for these analyses. Results are presented in Figure S3. Some bimodal dust distributions could potentially produce an AMEC-like cloud in scenarios in which the finer dust distribution has an effective radius of 10nm or smaller and high number densities of $100\text{-}1000\text{cm}^{-3}$. These scenarios are unrealistic (indicated by crosses in graphs), because small dust particles tend to coagulate, leading to larger effective radius of the smaller log-normal distribution³⁶.

We tried running mesoscale simulations using some of the (unrealistic due to coagulation) scenarios that could potentially be successful at producing the AMEC, according to the 0D model. The results are represented in the upper row of Figure S5, and they show that even in the best scenarios, the borders of the elongated tail tend to be blurry.

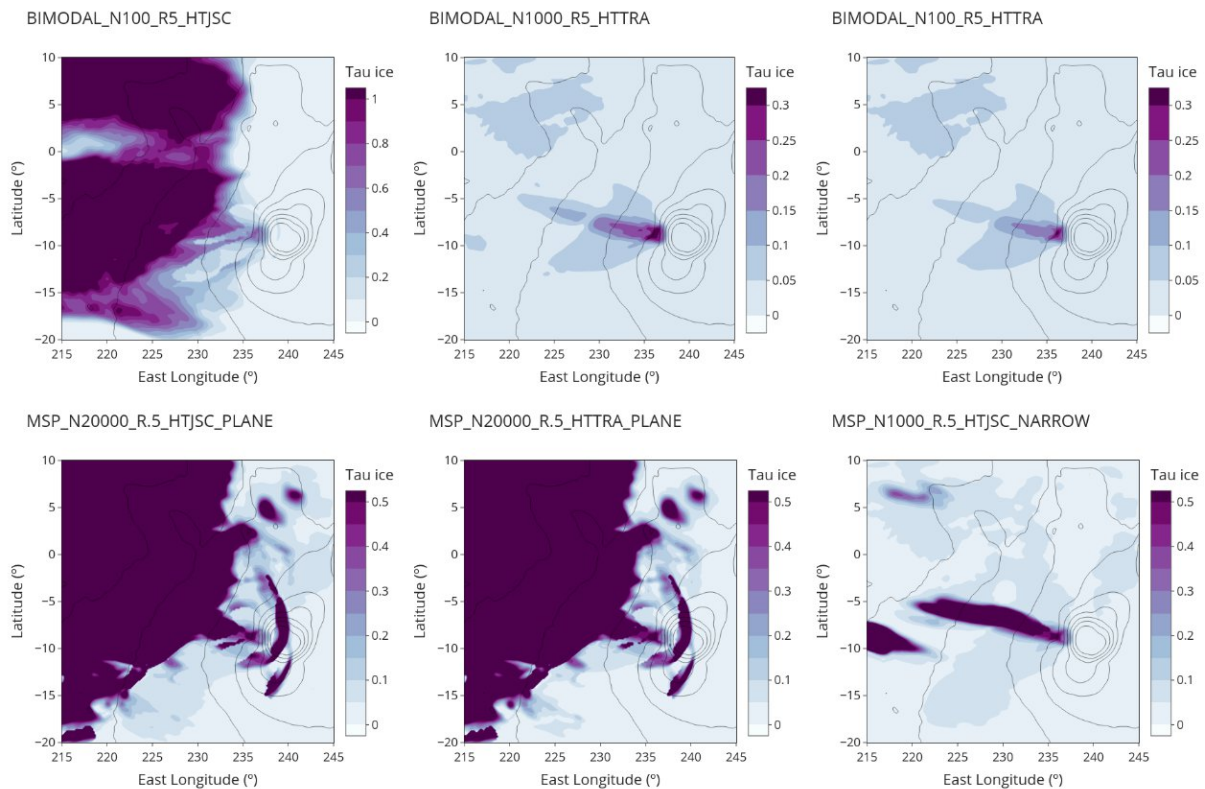


Figure S4. Optical depth obtained in mesoscale simulations for some scenarios of interest. See text and Extended Data Table S1 for details.

Another set of scenarios involving large number densities of particles with sizes around 0.5-10nm is given by dirty ice particles formed from meteoric smoke particles (MSPs), which have been analyzed for Mars by Plane et al.³⁹. They estimated in their figure 9 the size distribution of these particles as a function of the altitude. MSPs are originally tiny particles of 0.45nm, and they coagulate into larger particles. We extracted from Plane et al.³⁹ the size distribution at an altitude of 45km, which fits to a bimodal log-normal distribution with $N1=20000\text{cm}^{-3}$, $R1=0.45\text{nm}$, $N2=50\text{cm}^{-3}$, $R2=4\text{nm}$, with effective variances $v_{1eff} = 1$, $v_{2eff} = 0.1$ (figure S5, upper panel). The contact parameter of MSPs is expected to be high, realistic values^{46,69} would be around 0.99 - 1. We repeated the same approach as for the bimodal distribution, running a large number of simulations for a space of parameters that covers variations around the expected values (Table S2).

As in previous case, homogeneous nucleation was deactivated and only heterogeneous nucleation was allowed. For simplicity, the dust scenario was kept constant in these simulations, as a single log-normal mode with size $1\mu\text{m}$ and number density 0.1cm^{-3} , and the contact parameter of dust was set to HTJSC (the default value). Some of these scenarios produce points in the dispersion graph that fall within the area that correspond to a potentially realistic reproduction of the AMEC (figure S5, lower panel), however, these scenarios correspond to a low contact parameter of 0.95 (whereas a contact parameter close to 1 is to be expected^{6,69}) or small values of $N_2 < 5\text{cm}^{-3}$, much smaller than those predicted by Plane et al. Results of this experiment are shown in figure S4.

Parameter	Tested values	Number of values
Contact parameter	0.95, 0.99, 1	3
N1 (cm^{-3})	6000, 20000, 30000, 100000	4
R1 (nm)	0.45	1
N2 (cm^{-3})	1, 2, 5, 10, 50, 100, 200	7
R2 (nm)	2, 4, 6, 8	4

Table S2. Values considered in our exploration of the space of parameters for MSPs. The total amount of combinations is $336=3 \times 4 \times 1 \times 7 \times 4$. In this case, N1, and R1 refer to the number density and radius of the smaller mode. N2 and R2 refer to the larger mode, that forms by coagulation. The effective variances were set constant to $v_{1eff} = 1$, $v_{2eff} = 0.1$.

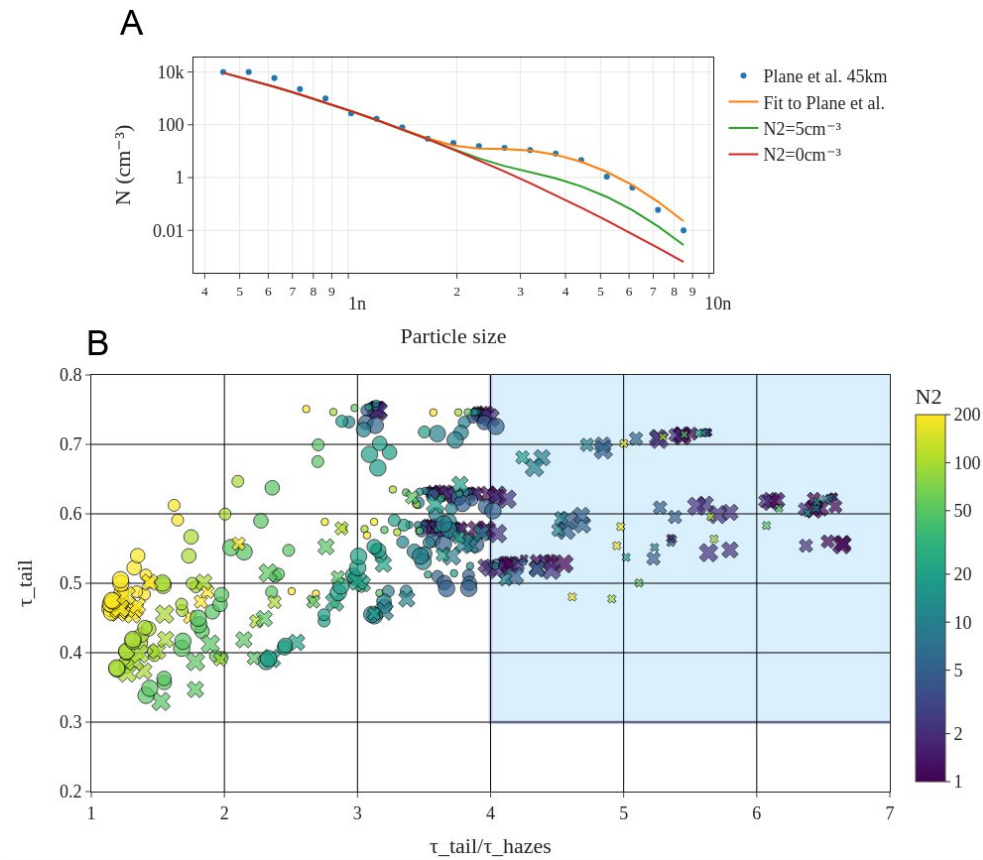


Figure S5. Results from 0D simulations with MSP, including only heterogeneous nucleation. (A) Size distribution of MSPs according to Planet et al. (Blue points) fit to it (orange), and specific variations of it (green,

red). (B) Simulation results equivalent to figure S3 but for MSPs, and covering the parameter space given in table S2. Unrealistic scenarios (Contact parameter 0.95, or $N_2 < 5 \text{ cm}^{-3}$. See text) are indicated by crosses.

We ran mesoscale simulations using some of the (unrealistic) scenarios that could potentially be successful at producing the AMEC, according to the 0D model. In the mesoscale model we cannot add bimodal distributions of MSPs, so we use a single mode. The red curve in upper panel in figure S5 is the MSP distribution employed in simulations MSP_N20000_R.5_HTTRA_PLANE and MSP_N20000_R.5_HTJSC_PLANE, which do not produce good reproductions of the AMEC. The results are represented in the lower row of Figure S5.

We made an additional intentionally extreme unrealistic test (MSP_N1000_R.5_HTJSC_NARROW), using a narrow distribution of MSPs including only particles of 0.5nm in radius, which is equivalent to assume that there is no coagulation and so larger particles are not available. This simulation was more successful at producing a realistic tail of the AMEC (figure S4).

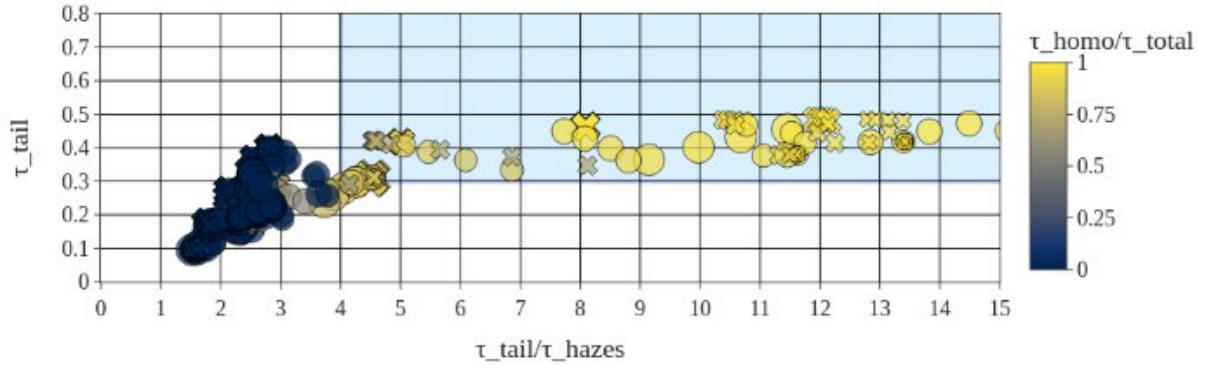
Finally, we repeated the 0D experiments considering the scenarios of bimodal dust distribution from table S1, but this time we activated homogeneous nucleation. Results are shown in Figure S6, and they show that homogeneous nucleation is the most reasonable explanation to the cloud that we observe.

In conclusion, only scenarios in which nucleation requires very high saturation ratios have the potential to produce a realistic AMEC. This is the case of homogeneous nucleation. Heterogeneous nucleation could mimic a similar behaviour only in scenarios that are not realistic, such as:

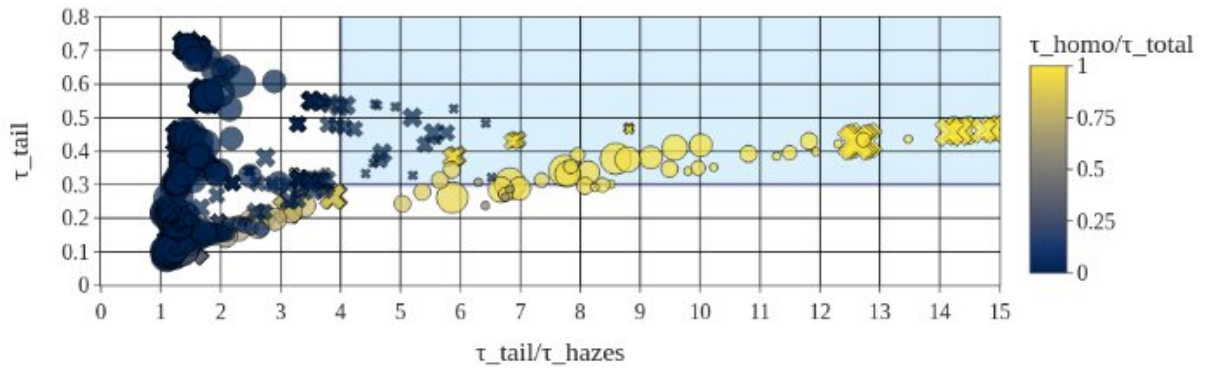
- A bimodal dust distribution in which the smaller mode comprises very small particles ($R_2 \leq 10 \text{ nm}$) in large number densities ($N_2 \geq 100 \text{ cm}^{-3}$) that would tend to disappear due to coagulation and are not supported by observations³⁶.
- The presence of MSPs that coagulate with much smaller efficiency than that predicted by Plane et al.³⁹, and with a contact parameter smaller than that expected for this kind of particles^{6,69}.

Therefore, these analyses rule out these alternative explanations for the AMEC, and reinforce the explanation involving homogeneous nucleation.

HTTRA



HTJSC



095

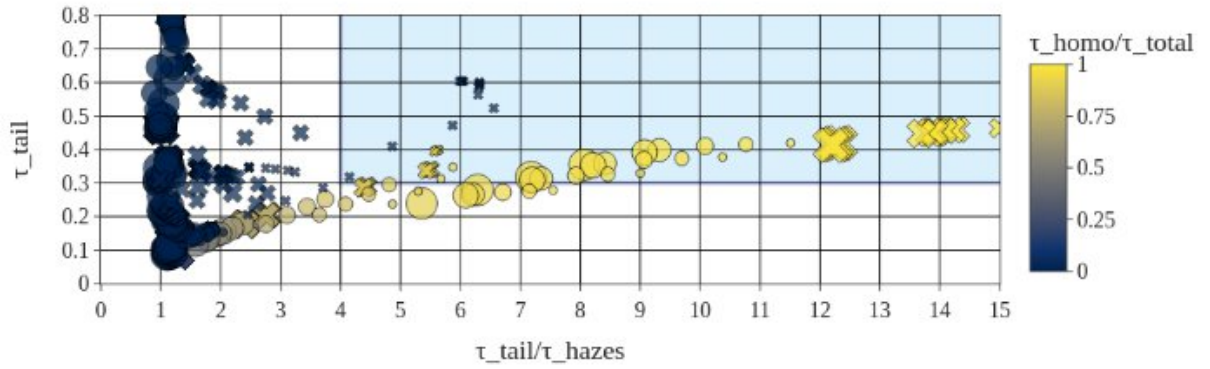


Figure S6. Simulation results with homogeneous nucleation. Same as figure S3 but with icehomogeneous nucleation activated. In this case the colour code indicates the fraction of optical depth that corresponds to homogeneously nucleated ice particles, supporting that only homogeneous nucleation is able to produce a realistic AMEC under realistic scenarios.

Supplementary Note S1. Contents of dataset in reference 44.

It contains all the materials required to reproduce this work: code, input files, and outputs. We describe here the folder structure. All details are given in README files.

- MESOSCALE: Everything related to mesoscale simulations and modeling.
 - CODE: Contains code files modified in the mesoscale model.
 - DATA
 - README
 - START_FILES.
 - README.TXT
 - Files containing initial and boundary conditions
 - Relevant configuration files
 - HETERO_REF: Outputs from simulation HETERO_REF.
 - HOMO_HTJSC: Outputs from simulation HOMO_HTJSC.
 - HOMO_OTHER: Outputs from all other simulations listed in table S1.
 - BIMODAL: Outputs from all bimodal simulations listed in EDF1.
 - MSP: Outputs from all MSP simulations listed in EDF1
- 0D_MICROPHYSICS: Everything related to 0D microphysics simulations and modeling.
 - CODE. Code of the model.
 - TIMESERIES: Timeseries of simulated lagrangian cells as extracted from the mesoscale model.
 - DATA
 - Discussion_S2. Datasets from Supplementary Discussion S2
 - EDF1. Datasets from Extended Data Figure 1
 - README