

Supporting Information for Multiscale atmospheric modeling suggests ammonia is necessary but not sufficient to explain new particle formation in the Colorado boundary layer

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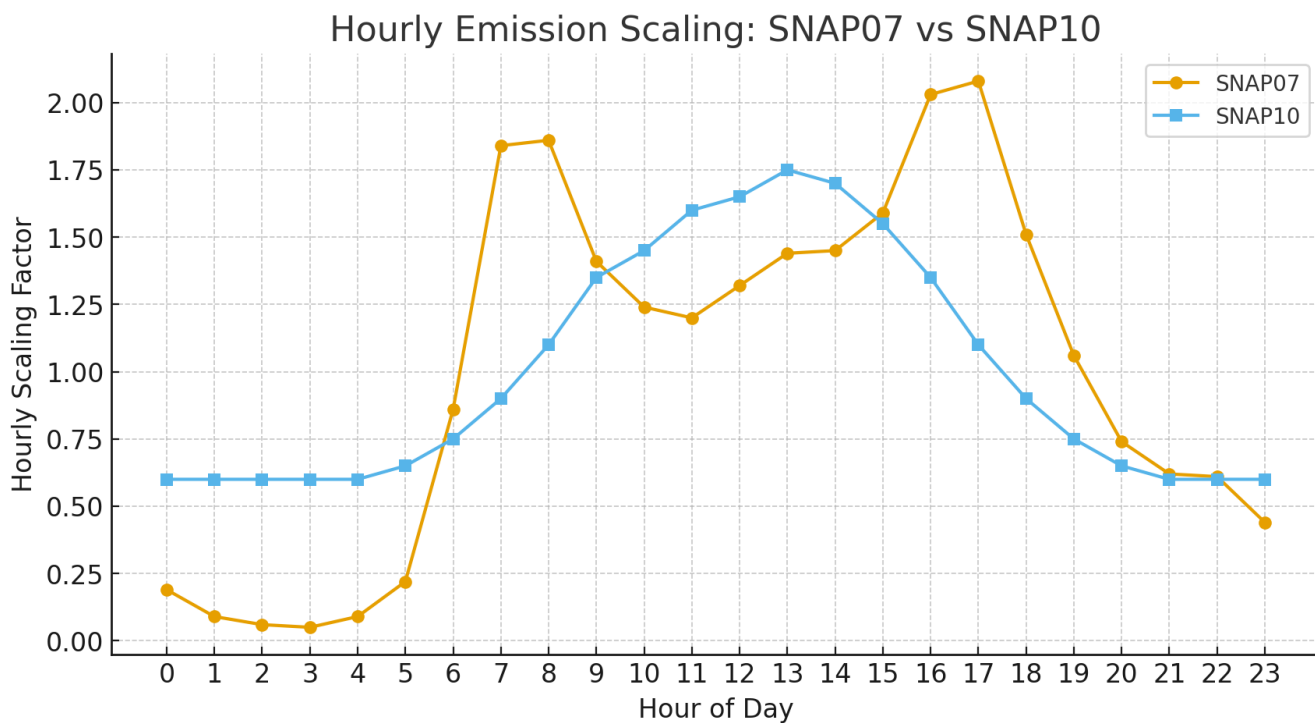


Figure S1. Hourly emission scaling cycles for our regional model for BC, OC, and NO_x fossil fuel emissions (SNAP07), and for NH₃ emissions (SNAP10), following Denier van der Gon et al. (2011).

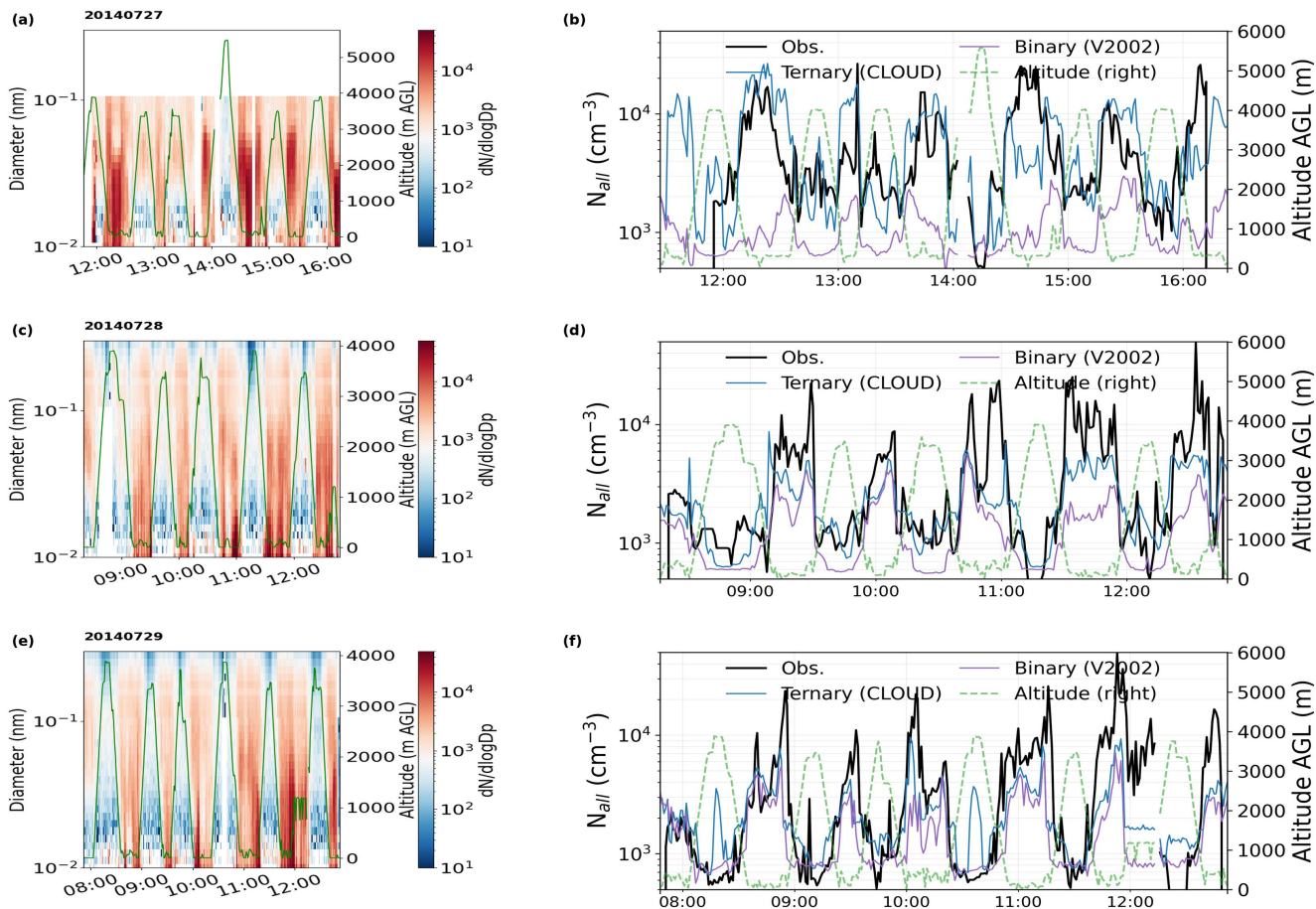


Figure S2. Observed size distributions and simulated and observed total aerosol number concentrations greater than 3 nm diameter during P-3 flights on 27, 28, 29 July 2014 . The P-3 LAS was not operational on 27 July.

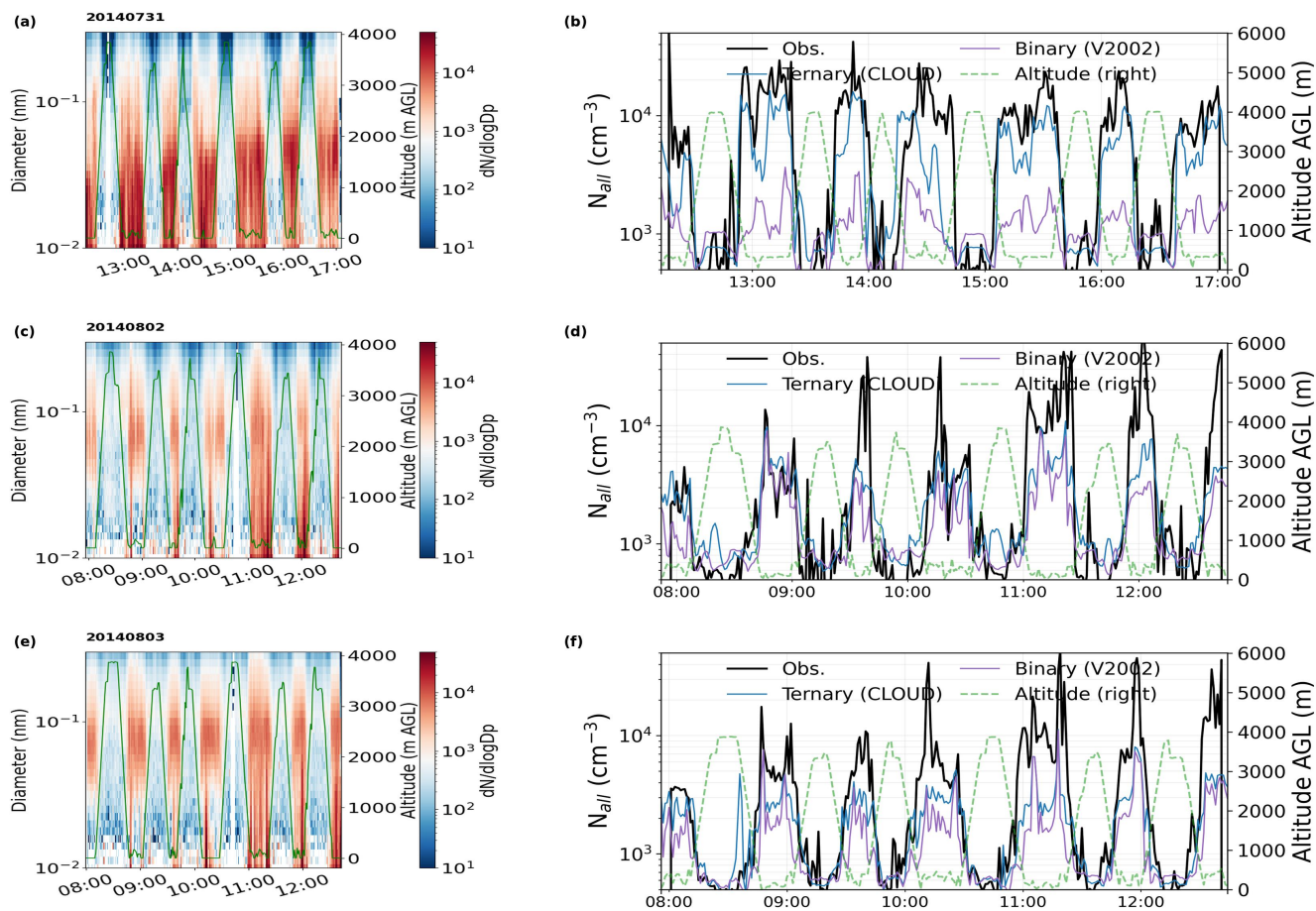


Figure S3. Observed size distributions and simulated and observed total aerosol number concentrations greater than 3 nm diameter during P-3 flights on 31 July, 2 and 3 August 2014.

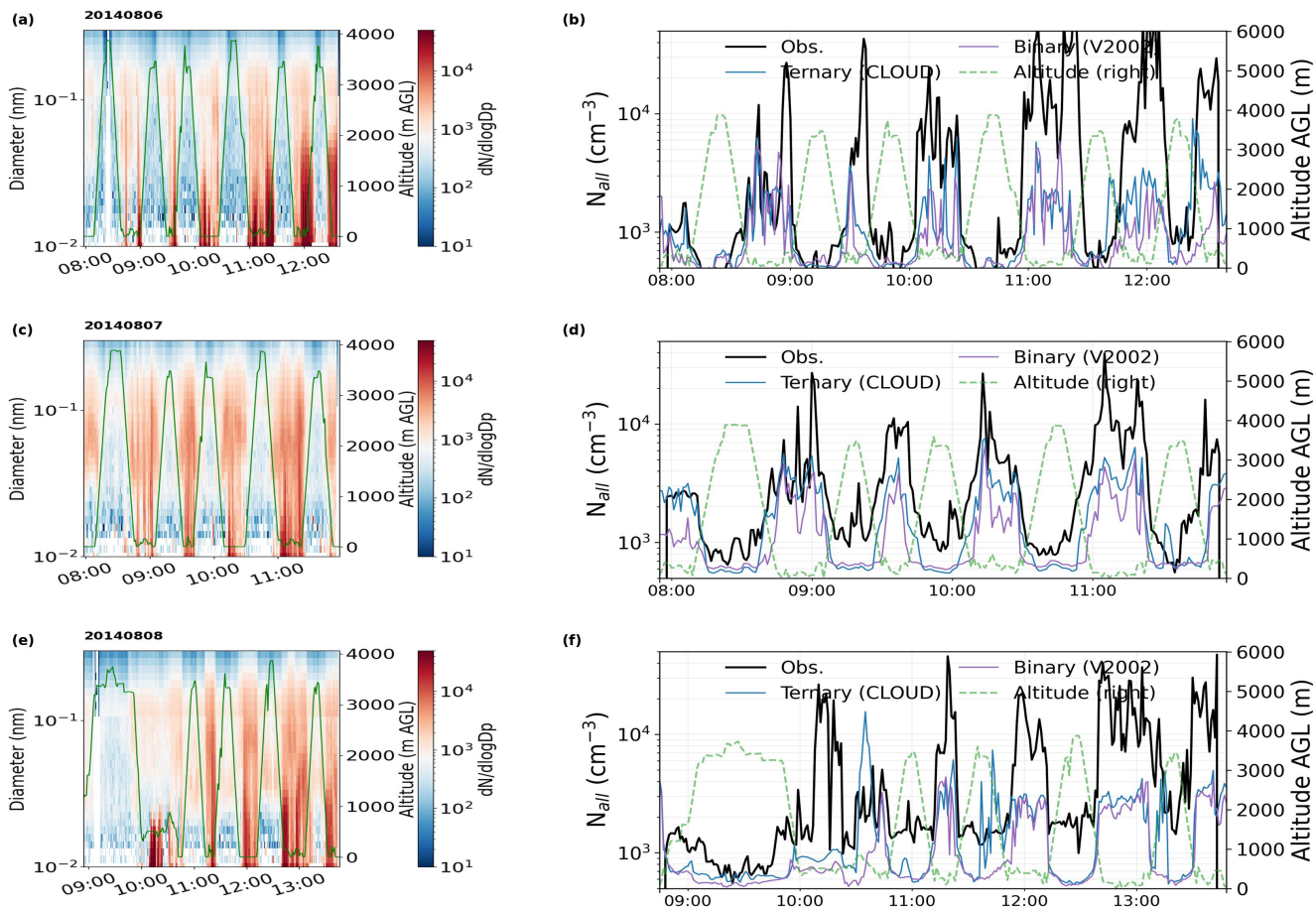


Figure S4. Observed size distributions and simulated and observed total aerosol number concentrations greater than 3 nm diameter during P-3 flights on 6, 7 and 8 August 2014. We only ran the simulation with the Dunne et al (2016) NPF parameterization on these days.

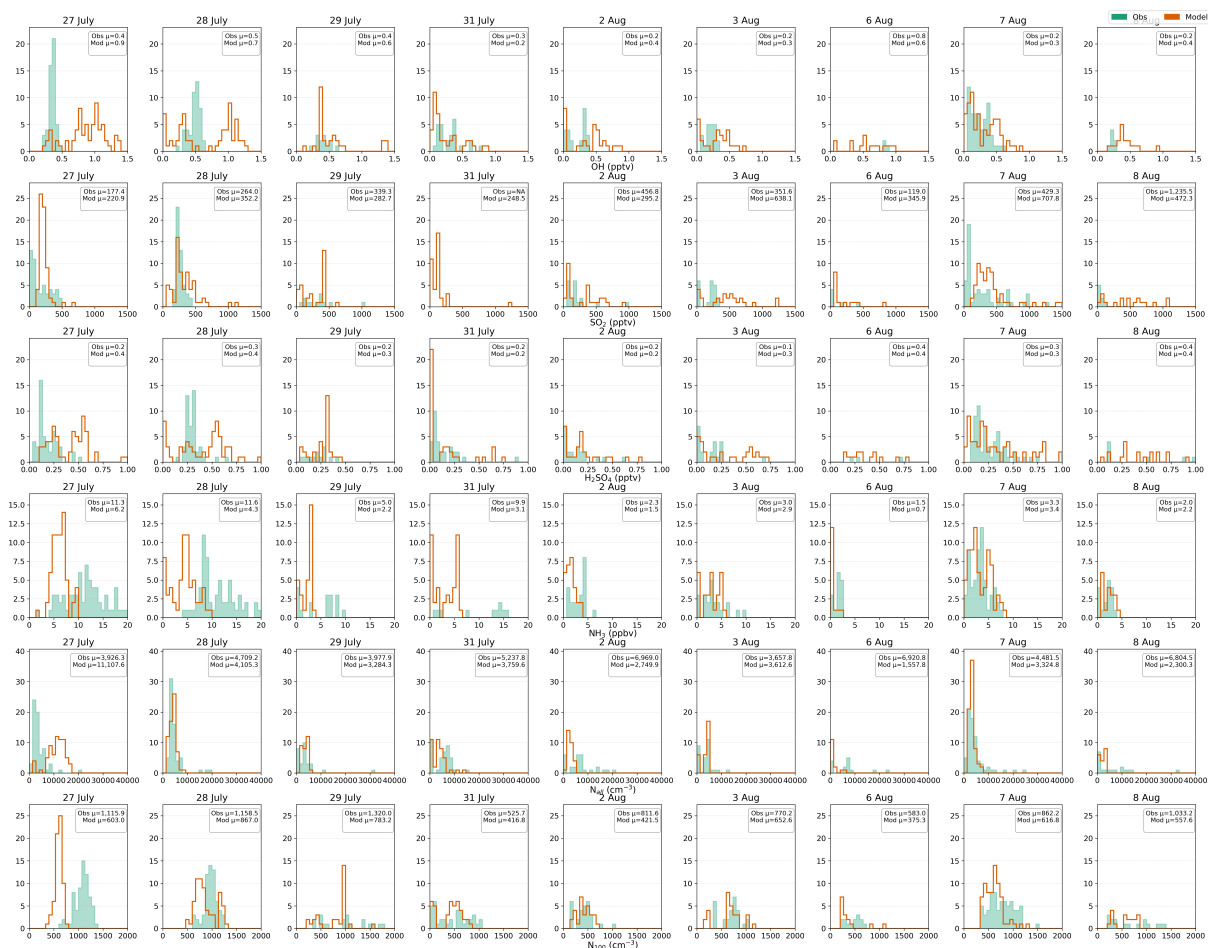


Figure S5. Species concentrations sampled by the C-130 aircraft, and simulated and interpolated onto the aircraft path, in a $0.4^\circ \times 0.4^\circ$ box around the Boulder Atmospheric Observatory tower, aggregated over all nine flights analyzed. All altitudes are included. The flight data and model output are sampled as one-minute averages, so the y axes of the histograms refer to the number of minutes for which the concentrations were equal to the value on the x axes. The SO₂ CIMS was not working on 31 July 2014 so observation data on that day is missing.

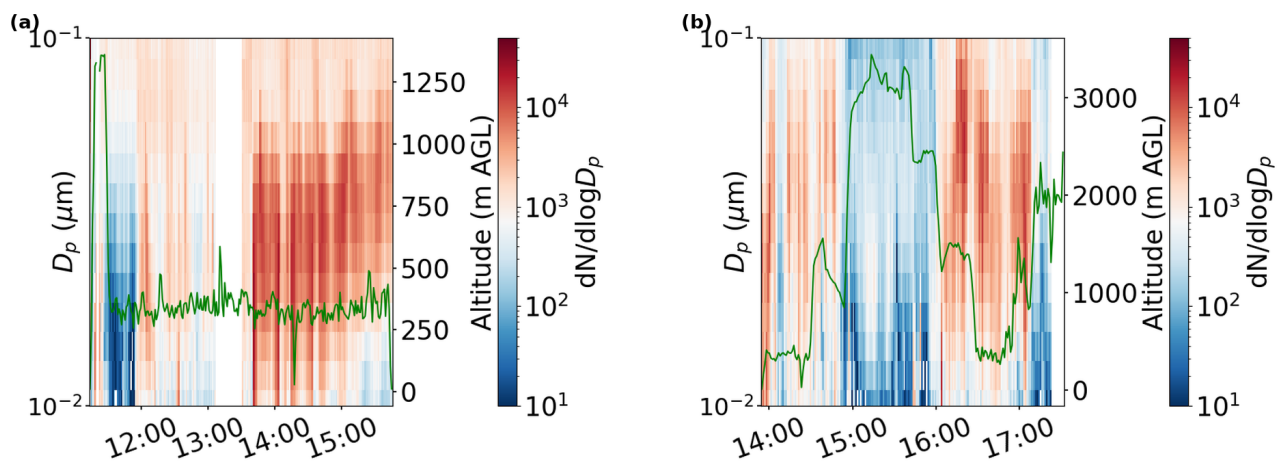


Figure S6. Size distributions observed by the C-130 aircraft's SMPS on 27 (left) and 31 (right) July 2014. There is evidence of NPF at the start of the flight and again after 1600 local time on 31 July, and after 1330 local time on 27 July.

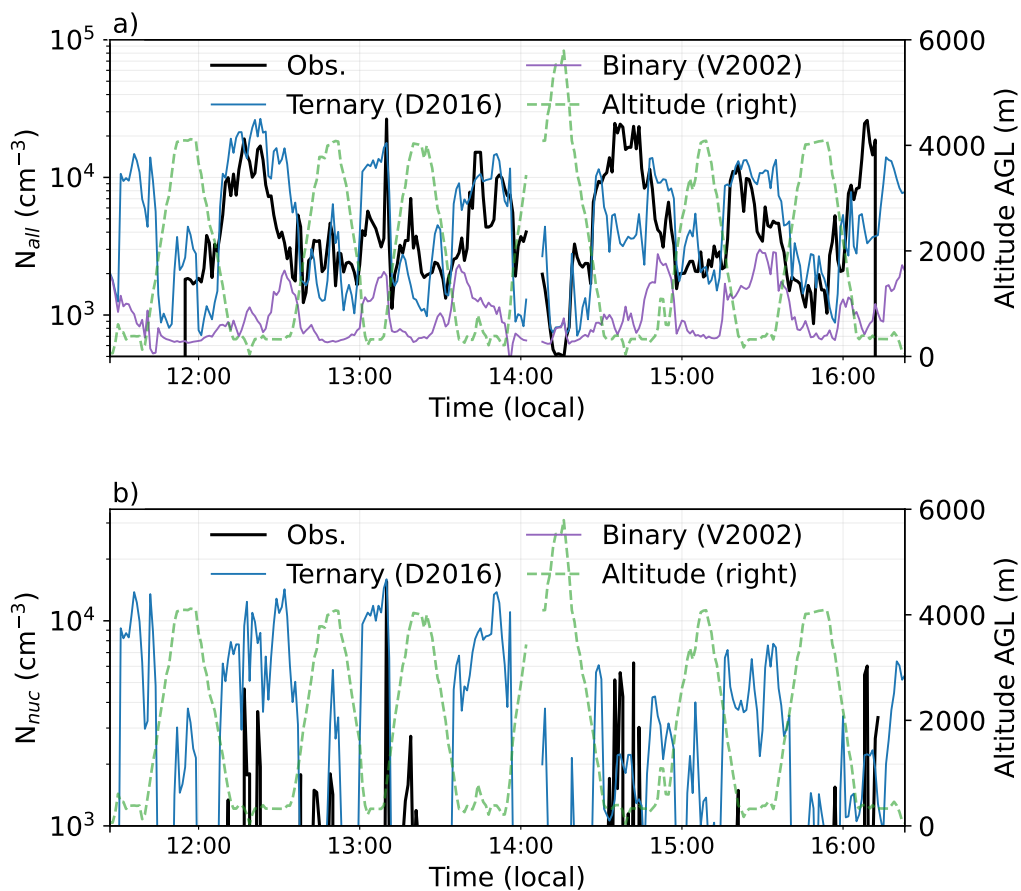


Figure S7. Total (a) and nucleation-mode (b) aerosol number concentrations observed by the P-3B on 27 July 2014 compared with the Unified Model using the Dunne et al. (2016) ternary and Vehkamäki et al. (2002) binary parameterizations. Altitude above ground level is shown on the secondary axis (green dashed). The aircraft instrument was operational between around 11:55 and 16:15.

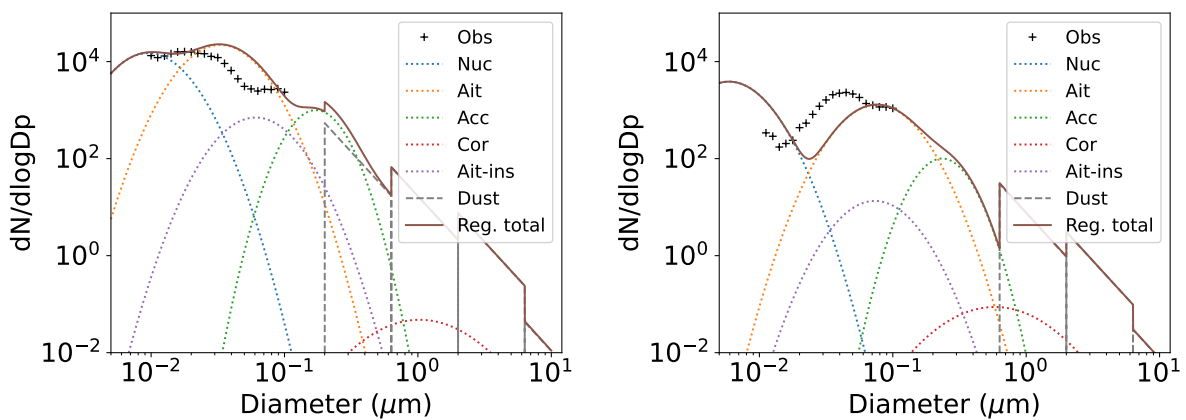


Figure S8. Size distributions observed by the P-3 aircraft's SMPS on 27 July 2014 near the surface (left) and at around 3500m above ground level (right). There is evidence of NPF at the start of the flight and again after 1600 local time on 31 July, and after 1330 local time on 27 July.

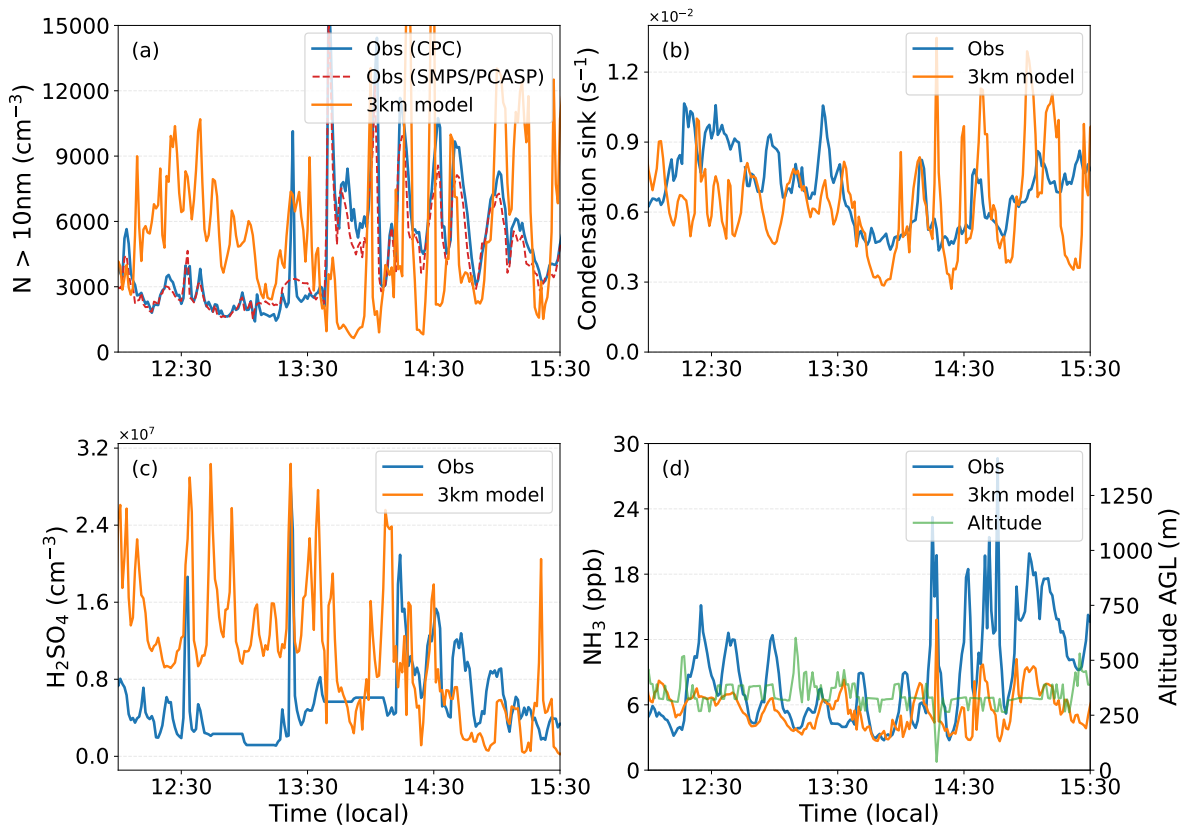


Figure S9. Particle number concentration a), condensation sink (b), sulfuric acid c), and ammonia d) concentrations observed by the C-130 aircraft, and simulated, on 27 July 2014.

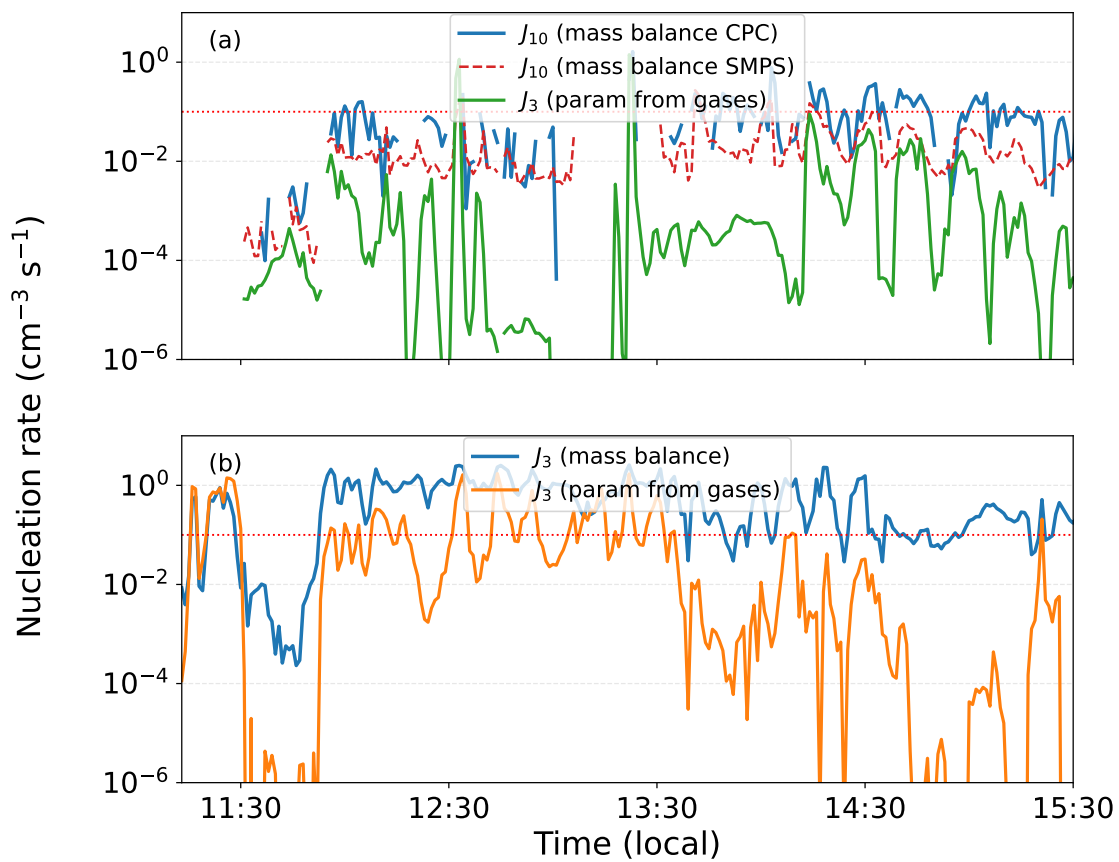


Figure S10. NPF rates calculated from a mass balance (see text) and calculated from observed gas concentrations using the Dunne et al (2016) parameterization (a), and the corresponding figure for the UM simulations (b), on 27 July 2014.

References

- Denier van der Gon, H., Hendriks, C., Kuenen, J., Segers, A., and Visschedijk, A.: Description of current temporal emission patterns and sensitivity of predicted AQ for temporal emission patterns. TNO Rep. EU FP7 MACC Deliverable Rep. D_DEMIS_1. 3, 22 pp, 2011.
- Dunne, E. M., Gordon, H., K^oErten, A., Almeida, J., Duplissy, J., Williamson, C., Ortega, I. K., Pringle, K. J., Adamov, A., Baltensperger, U., Barmet, P., Benduhn, F., Bianchi, F., Breitenlechner, M., Clarke, A., Curtius, J., Dommen, J., Donahue, N. M., Ehrhart, S., Flagan, R. C., Franchin, A., Guida, R., Hakala, J., Hansel, A., Heinritzi, M., Jokinen, T., Kangasluoma, J., Kirkby, J., Kulmala, M., Kupc, A., Lawler, M. J., Lehtipalo, K., Makhmutov, V., Mann, G., Mathot, S., Merikanto, J., Miettinen, P., Nenes, A., Onnela, A., Rap, A., Reddington, C. L. S., Riccobono, F., Richards, N. A. D., Rissanen, M. P., Rondo, L., Sarnela, N., Schobesberger, S., Sengupta, K., Simon, M., Sipil^o, M., Smith, J. N., Stozkhov, Y., Tom^o, A., Trostl, J., Wagner, P. E., Wimmer, D., Winkler, P. M., Worsnop, D. R., and Carslaw, K. S.: Global atmospheric particle formation from CERN CLOUD measurements, *Science*, 354, 1119–1124, <https://doi.org/10.1126/science.aaf2649>, 2016.
- Vehkamäki, H., Kulmala, M., Napari, I., Lehtinen, K. E. J., Timmreck, C., Noppel, M., and Laaksonen, A.: An improved parameterization for sulfuric acid–water nucleation rates for tropospheric and stratospheric conditions, *Journal of Geophysical Research*, 107, 4622, <https://doi.org/10.1029/2002JD002184>, 2002.