Supplementary Material

Demonstration of Time Dependency in Model Simulations

Supplementary Figure 1 demostrates the large amount of simulation time required by the parcel model in order to activate a fraction of large aerosol, 1500 nm.

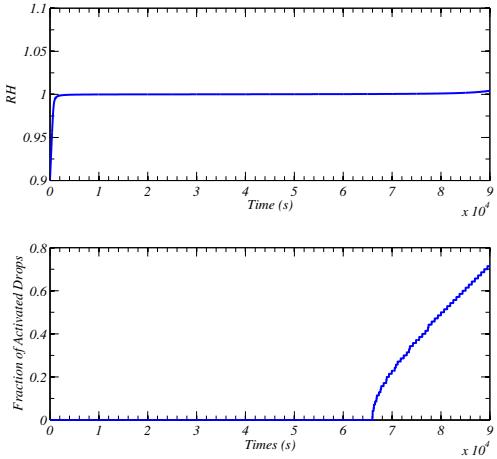

Fig. 1: Time series of peak RH (top panel) and fraction of activated drops (bottom panel) as calculated by ACPIM with initial conditions described in Table 1, aerosol number concentration 500 cm^{-3}, and mean aerosol diameter 1500 nm.

Single-Mode Experiment

Supplementary Figure 2 shows that the preformance of the three parameterisations is generally good in single lognormal aerosol size distribution of small aerosol ($5 \leq d_m \leq 250$ nm) at high number concentration, 10000 cm^{-3}. However there is a tendency to underestimate the fraction of activated drops.

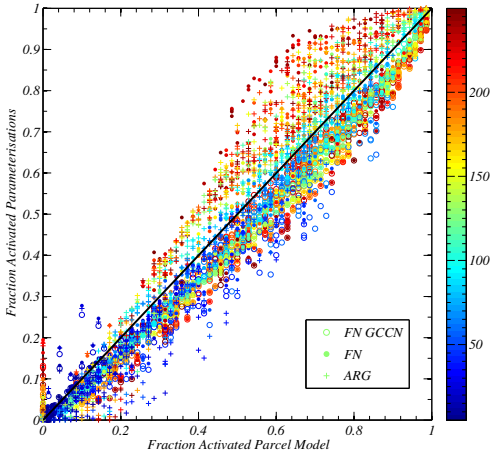

Fig. 2: Results from 1500 runs with 1 lognormal mode of ammonium sulphate aerosol, number concentration of 10000 cm^{-3}. The values for the following variables were randomly chosen within the stated ranges: $0.2 \leq \ln\sigma \leq 0.8$; median diameter, $5 \leq d_m \leq 250$ nm; updraft velocity, $0.01 \leq w \leq 10$ m s^{-1}. Initial conditions are detailed in Table 1.

Supplementary Figure 3 shows that the three parameterisations highly overestimate the fraction of activated drops in single lognormal aerosol size distribution of large aerosol ($250 \leq d_m \leq$

2000 nm) at low number concentration, 100 cm^{-3}.

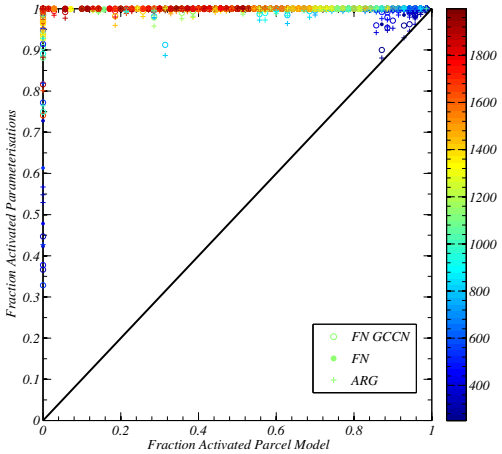

Fig. 3: Results from 1500 runs with 1 lognormal mode of ammonium sulphate aerosol, number concentration of 100 cm^{-3}. Symbols are coloured by median aerosol diameter. The values for the following variables were randomly chosen within the stated ranges: $0.2 \leq \ln\sigma \leq 0.8$; median diameter, $250 \leq d_m \leq 2000$ nm; updraft velocity, $0.01 \leq w \leq 10$ m s^{-1}. Initial conditions are detailed in Table 1.

Supplementray Figures 4 and 5 show the same results as Figure 2 with data points coloured coded by $\ln\sigma$ and aerosol number concentration respectively. There is no obvious relationship between the fraction of activated drops estimated by the parameterisations and variables mentioned.

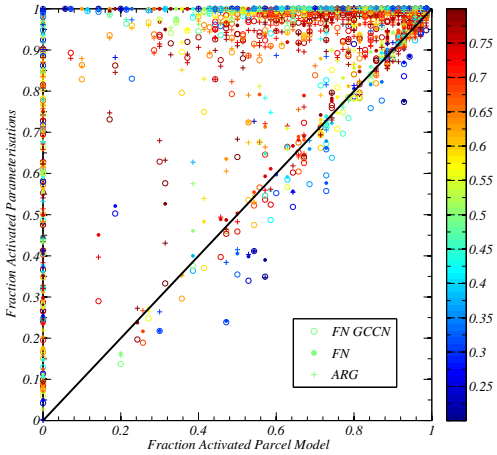

Fig. 4: Results from 1500 runs with 1 lognormal mode of ammonium sulphate aerosol and randomly sampled variables values as detailed in Table 2 and initial conditions described in Table 1. Symbols are coloured by $\ln\sigma$, see colour bar.

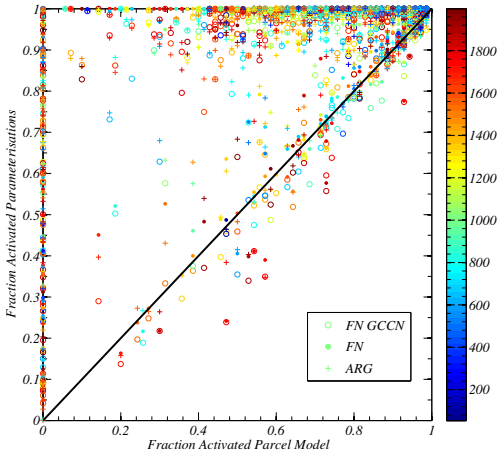

Fig. 5: Results from 1500 runs with 1 lognormal mode of ammonium sulphate aerosol and randomly sampled variables values as detailed in Table 2 and initial conditions described in Table 1. Symbols are coloured by aerosol number concentration (number cm^{-3}), see colour bar.

Dual-Mode Experiment

To avoid extreme conditions of very high concentrations of large or small aerosol the parameter space has been limited in the following cases. The range of updraft velocity has also been limited so that within the simulation time of the parcel model the "effective height" of the rising air parcel is more realistic, i.e between 0.4 km and 10.4km, where clouds form.

Supplementray Figure 6 shows that ARG underestimates peak supersaturation while FN GCCN tends to overestimate peak supersaturation.

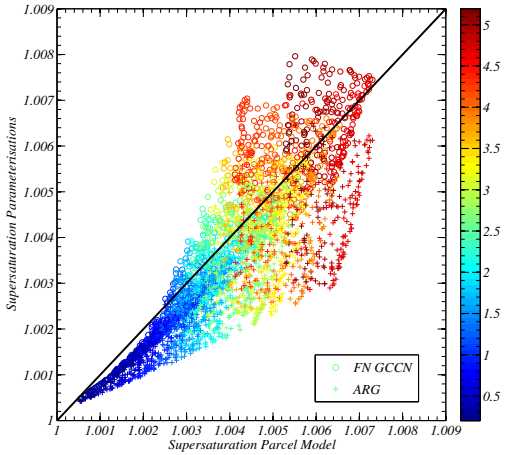

Fig. 6: Results for peak RH from 1500 runs with bimodal aerosol size distributions. Only the median diameter of aerosol in the 2nd mode and updraft velocity were changed between runs within the ranges of $100 \leq d_m \leq 800$ nm and $0.2 \leq w \leq 5.2$ m s^{-1} respectively. Symbols are coloured by updraft velocity, see colourbar. For initial conditions see Table 1.

Using a smaller parameter space improves the results from the three parameterisations as can be seen in Supplementary Figure 7.

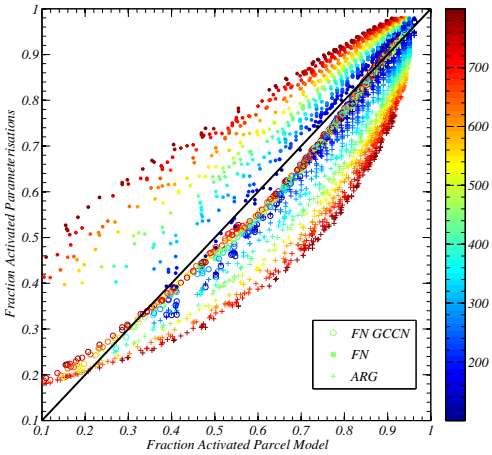

Fig. 7: Results from 1500 runs with bimodal aerosol size distributions, 500 cm^{-3} in the first mode and 100 cm^{-3} in the second. Only the median diameter of aerosol in the second mode and updraft velocity were changed between runs, within the ranges of $100 \leq d_m \leq 800$ nm and $0.2 \leq w \leq 5.2$ m s^{-1} respectively. Symbols are coloured by median diameter (nm) of the second mode, see colourbar. For initial conditions see Table 1.

Using a longer runtime in the parcel model the feature in Figure 3 at 0.16 fraction of activated drops is less pronounced –see Supplementary Figure 8, demostrating that the parameterisations are activing aerosol that otherwise would not grow to their critical diameters.

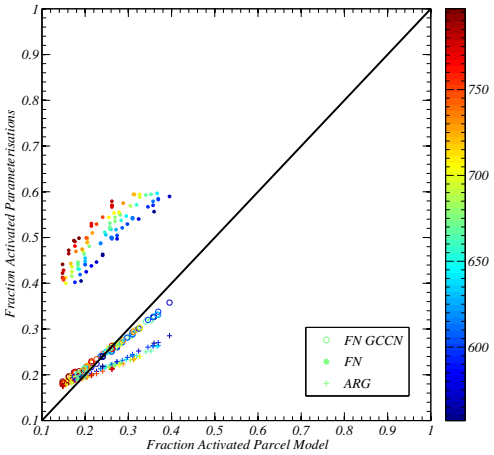

Fig. 8: Results from 100 runs with bimodal aerosol size distributions, 500 cm^{-3} in first mode and 100 cm^{-3} in the second mode, using a runtime of 8000 s in the parcel model. Only the median diameter of aerosol in the 2nd mode and updraft velocity were changed between runs within the ranges of $550 \leq d_m \leq 800$ nm and $0.2 \leq w \leq 0.5$ m s^{-1} respectively. Symbols are coloured by median diameter (nm) of the second mode, see colourbar. For initial conditions see Table 1.