Electronic Supplementary Material (ESI) for Environmental Science: Atmospheres.

Single particle measurements of mixing between mimics for biomass burning and aged secondary organic aerosols

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Supporting Information:
15 pages
14 Figures

Supplemental Figures:

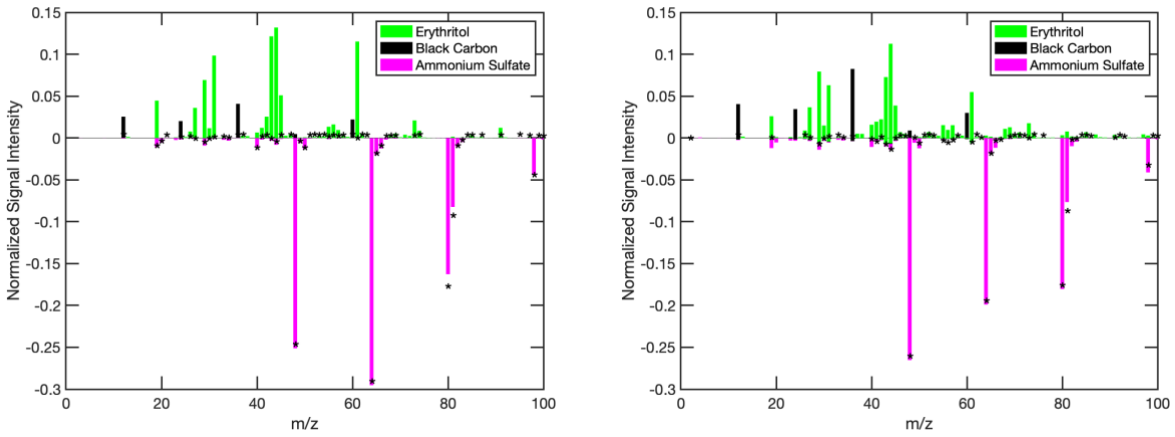

Figure S1: Aggregated mass spectra of particle populations from the start (left) to end (right) of mixing event. We added particles with volatile erythritol (green) coating black carbon (black) into a chamber containing particles with ammonium sulfate (magenta) at 20 ºC and 5 % RH. Over the course of the experiment the erythritol signal decreases relative to the black carbon signal, made evident by the decreases in the green *m/z* sticks relative to the black m/z sticks. However, there is very little observed increase in corresponding *m/z* sticks in the ammonium sulfate population, confirming that erythritol does not condense onto ammonium sulfate. The asterisks represent the residual from the linear combination.

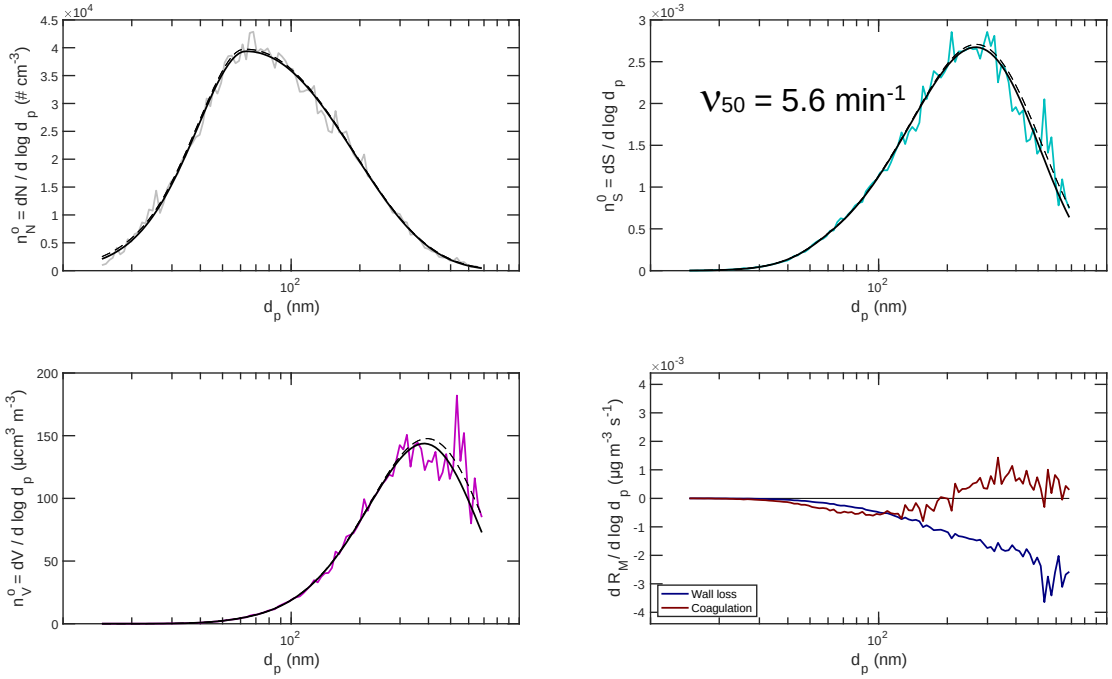

Figure S2: Particle size distributions and gain/loss rates from the mixing experiment conducted with the erythritol/black carbon and ammonium sulfate populations at 20 ºC and 5 % RH. These panels represent data collected immediately after introducing the sucrose/black carbon population into the chamber, when the particle concentration of both populations together would be the highest. The top left panel shows a particle number distribution with a size mode between 60-90 nm and a total particle concentration of ~30,000 # cm^{-3}. The top right panel shows a particle surface area distribution with a size mode between 200-300 nm and particle surface area concentration of ~$1.9x10^{-3}$ m^2 m^{-3}. The bottom left panel shows the particle volume distribution with a size mode between 300-400 nm and a particle volume concentration of ~80 µcm^3 m^{-3}. The bottom right panel shows the calculated particle gain/loss rates in terms of particle mass for coagulation and wall losses. This experiment had a total coagulation loss rate of ~5,400 # cm^{-3} hr^{-1}, however the resulting change in particle mass in particles that are seen by the AMS (≥100 nm) is ~$9x10^{-6}$ µg cm^{-3} hr^{-1}. Thus, coagulation is not a major source of mixing in this system.

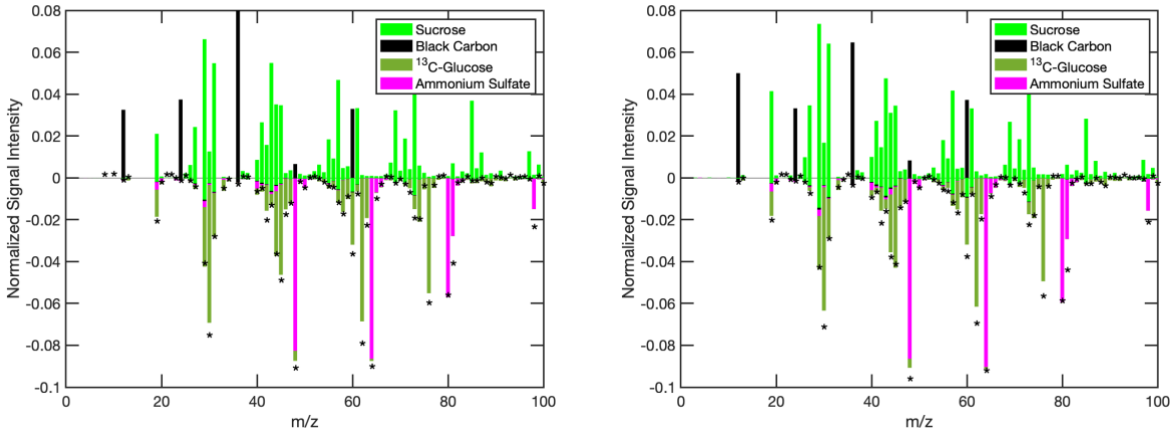

Figure S3: Aggregated mass spectra of particle populations from the start (left) to end (right) of mixing event. We added particles with non-volatile sucrose (green) coating black carbon (black) into a chamber containing particles with non-volatile ^{13}C-glucose (olive) coating ammonium sulfate (magenta) at 20 ºC and 5 % RH. Over the course of the experiment the sucrose signal is stable relative to the black carbon signal, made evident by the changes in the green *m/z* sticks, but no observable relative decrease in signal intensity compared to the black m/z sticks. There is also very little observed increase in corresponding *m/z* sticks in the ^{13}C-glucose/ammonium sulfate population, other than cross-talk between the spectra. This confirms that increasing organic signal in non-volatile sugar populations over the course of these mixing experiments indicates uptake of erythritol vapors into the organic fraction of the opposite population. The asterisks represent the residual from the linear combination calculation.

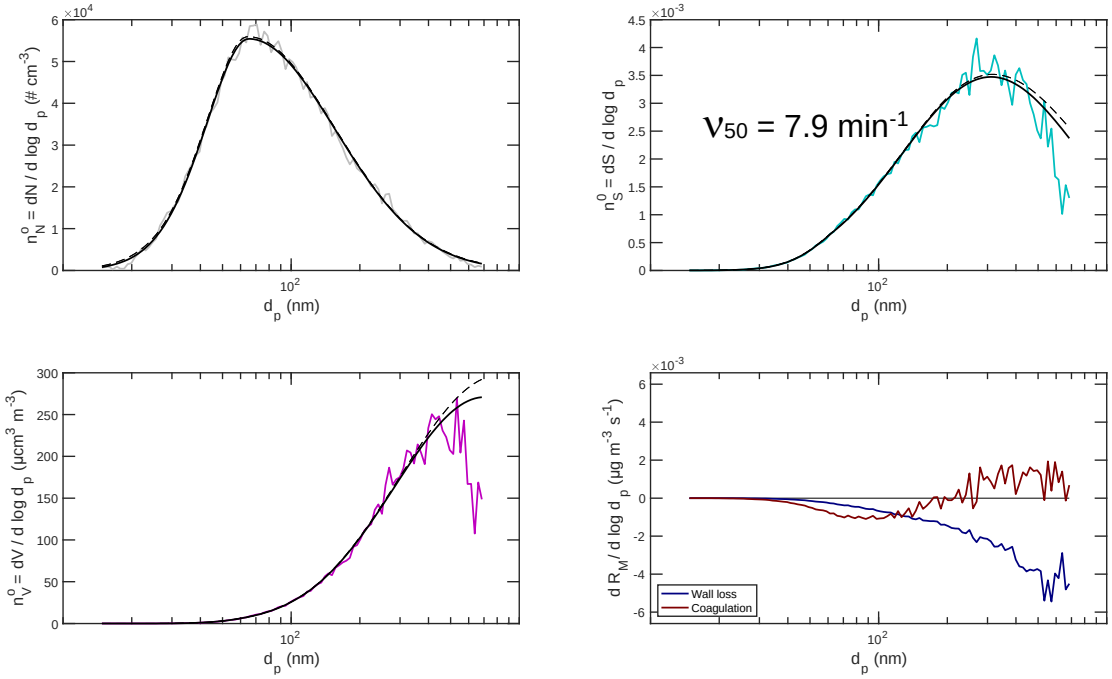

Figure S4: Particle size distributions and gain/loss rates from the mixing experiment conducted with the sucrose/black carbon and ^{13}C-glucose/ammonium sulfate populations at 20 ºC and 5 % RH. These panels represent data collected immediately after introducing the sucrose/black carbon population into the chamber, when the particle concentration of both populations together would be the highest. The top left panel shows a particle number distribution with a size mode just under 100 nm and a total particle concentration of ~40,000 #/cm^3. The top right panel shows a particle surface area concentration with a size mode around 250 nm and particle surface area concentration of 2.6e-3 m^2/m^3. The bottom left panel shows the particle volume distribution with a size mode between 600-700 nm and a particle volume concentration of ~121 µcm^3 m^{-3}. The bottom right panel shows the calculated particle gain/loss rates in terms of particle mass for coagulation and wall losses. This experiment had a total coagulation loss rate of ~7,200 # cm^{-3} hr^{-1}, however the resulting change in particle mass in particles that are seen by the AMS (≥100 nm) is ~1.6x10^{-5} µg cm^{-3} hr^{-1}. Thus, coagulation is not a major source of mixing in this system.

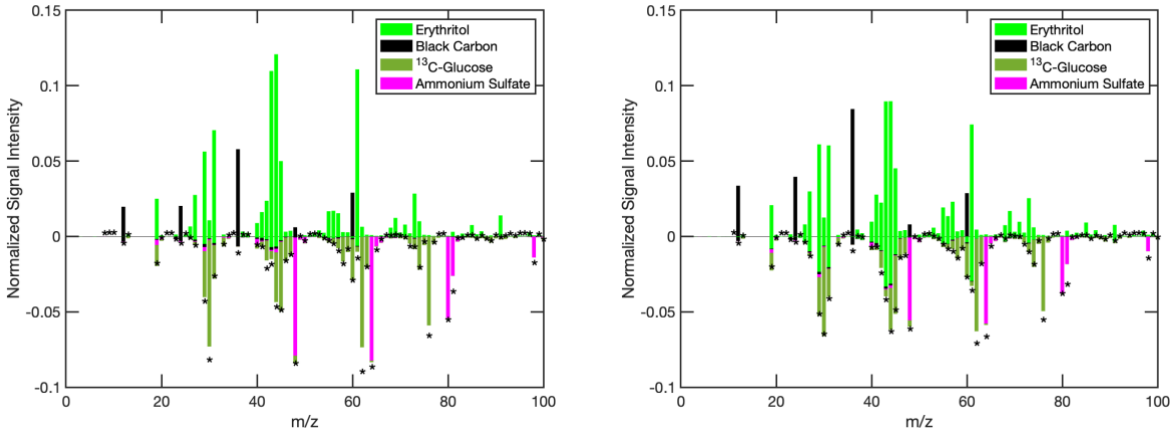

Figure S5: Aggregated mass spectra of particle populations from the start (left) to end (right) of mixing event. We added particles with volatile erythritol (green) coating black carbon (black) into a chamber containing particles with non-volatile ^{13}C-glucose (olive) coating ammonium sulfate (magenta) at 20 ºC and 5 % RH. Over the course of the experiment the erythritol signal decreases relative to the black carbon signal, made evident by the decreases in the green *m/z* sticks relative to the black m/z sticks. There is also an observed increase in corresponding *m/z* sticks in the ^{13}C-glucose/ammonium sulfate population. This suggests that mixing is generally uninhibited for this system at these conditions. The asterisks represent the residual from the linear combination calculation.

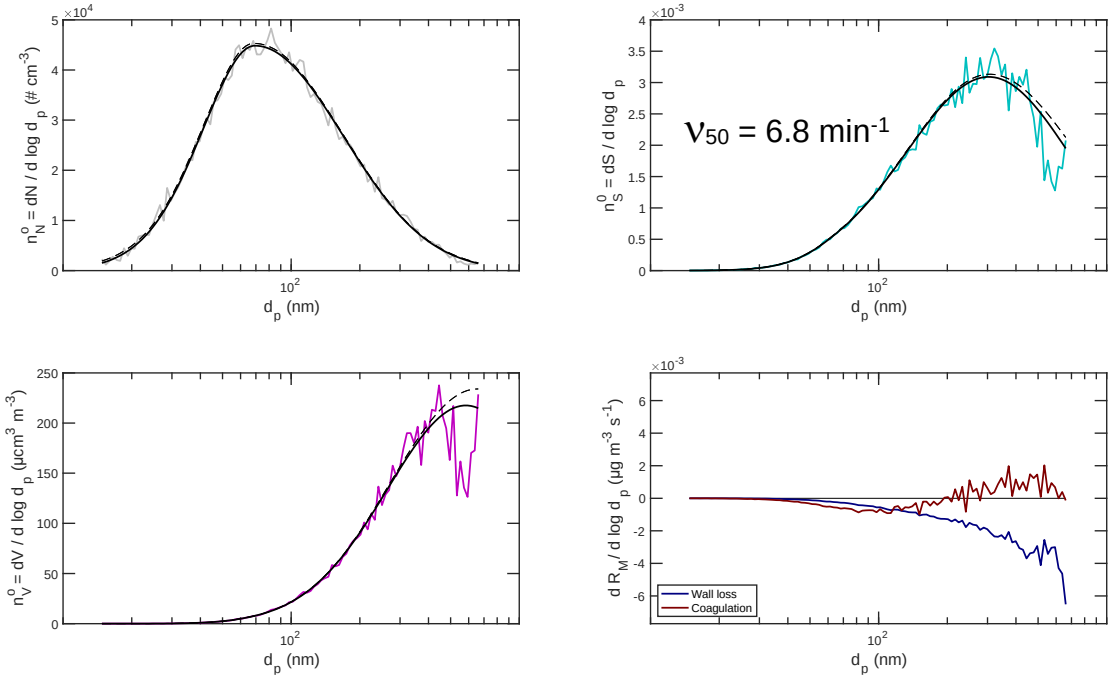

Figure S6: Particle size distributions and gain/loss rates from the mixing experiment conducted with the erythritol/black carbon and ^{13}C-glucose/ammonium sulfate populations at 20 ºC and 5 % RH. These panels represent data collected immediately after introducing the erythritol/black carbon population into the chamber, when the particle concentration of both populations together would be the highest. The top left panel shows a particle number distribution with a size mode just under 100 nm and a total particle concentration of ~20,000 #/cm^3. The top right panel shows a particle surface area concentration with a size mode around 250 nm and particle surface area concentration of 1.4e-3 m^2/m^3. The bottom left panel shows the particle volume distribution with a size mode between 500-600 nm and a particle volume concentration of ~100 µcm^3 m^{-3}. The bottom right panel shows the calculated particle gain/loss rates in terms of particle mass for coagulation and wall losses. This experiment had a total coagulation loss rate of ~6,300 # cm^{-3} hr^{-1}, however the resulting change in particle mass in particles that are seen by the AMS (≥100 nm) is ~1.2x10^{-5} µg cm^{-3} hr^{-1}. Thus, coagulation is not a major source of mixing in this system.

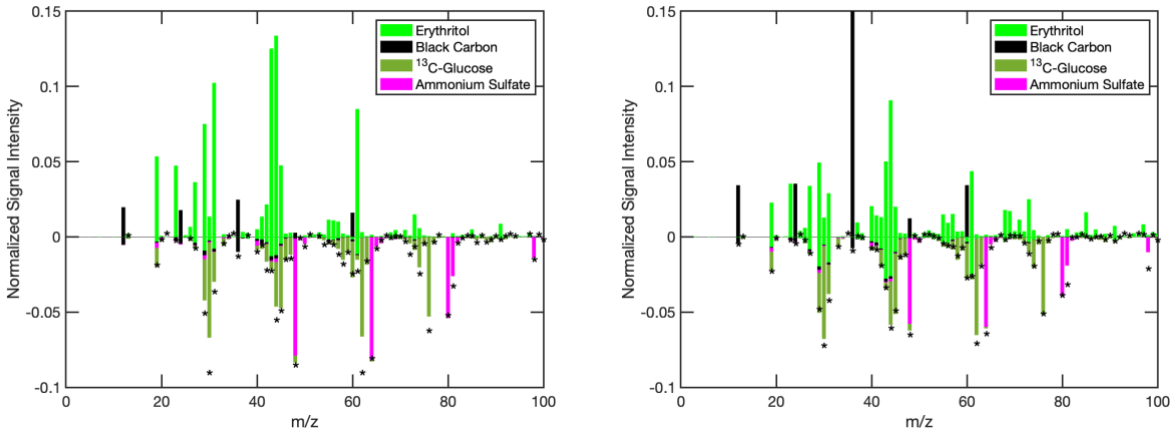

Figure S7: Aggregated mass spectra of particle populations from the start (left) to end (right) of mixing event. We added particles with volatile erythritol (green) coating black carbon (black) into a chamber containing particles with non-volatile ^{13}C-glucose (olive) coating ammonium sulfate (magenta) at 10 ºC and 5 % RH. Over the course of the experiment the erythritol signal decreases relative to the black carbon signal, made evident by the decreases in the green *m/z* sticks relative to the black m/z sticks. There is also an observed increase in corresponding *m/z* sticks in the ^{13}C-glucose/ammonium sulfate population. This suggests that mixing is generally uninhibited for this system at these conditions. The asterisks represent the residual from the linear combination calculation.

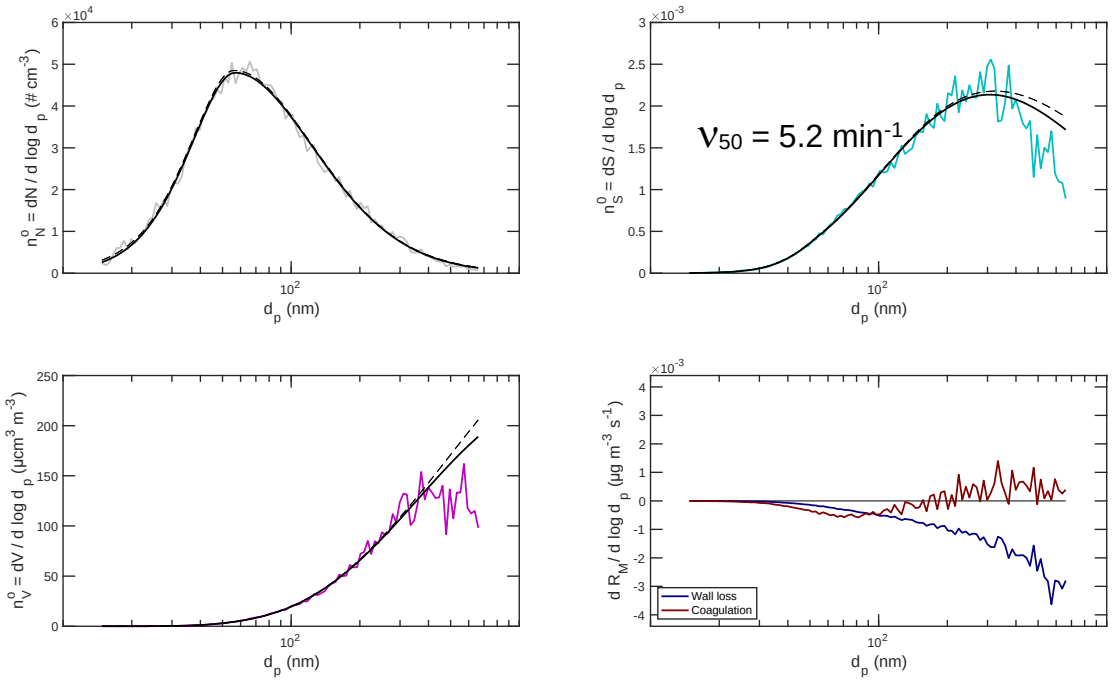

Figure S8: Particle size distributions and gain/loss rates from the mixing experiment conducted with the erythritol/black carbon and ^{13}C-glucose/ammonium sulfate populations at 10 ºC and 5 % RH. These panels represent data collected immediately after introducing the erythritol/black carbon population into the chamber, when the particle concentration of both populations together would be the highest. The top left panel shows a particle number distribution with a size mode just under 100 nm and a total particle concentration of ~35,000 #/cm^3. The top right panel shows a particle surface area concentration with a size mode around 250 nm and particle surface area concentration of 1.7e-3 m^2/m^3. The bottom left panel shows the particle volume distribution with a size mode between 300-500 nm and a particle volume concentration of ~100 µcm^3 m^{-3}. The bottom right panel shows the calculated particle gain/loss rates in terms of particle mass for coagulation and wall losses. This experiment had a total coagulation loss rate of ~7,200 # cm^{-3} hr^{-1}, however the resulting change in particle mass in particles that are seen by the AMS (≥100 nm) is ~9x10^{-6} µg cm^{-3} hr^{-1}. Thus, coagulation is not a major source of mixing in this system.

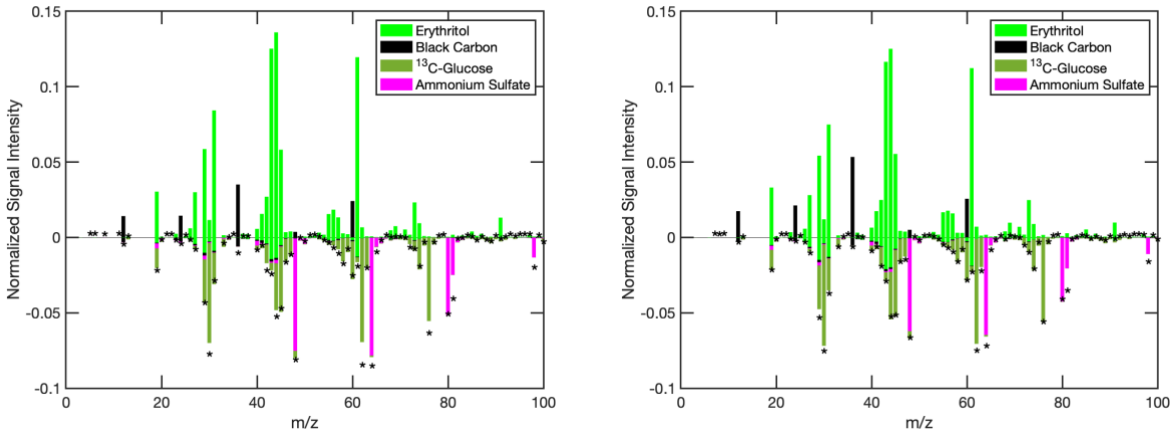

Figure S9: Aggregated mass spectra of particle populations from the start (left) to end (right) of mixing event. We added particles with volatile erythritol (green) coating black carbon (black) into a chamber containing particles with non-volatile ^{13}C-glucose (olive) coating ammonium sulfate (magenta) at 10 ºC and 90 % RH. Over the course of the experiment the erythritol signal decreases relative to the black carbon signal, made evident by the decreases in the green *m/z* sticks relative to the black m/z sticks. There is also an observed increase in corresponding *m/z* sticks in the ^{13}C-glucose/ammonium sulfate population. This suggests that mixing is generally uninhibited for this system at these conditions. The asterisks represent the residual from the linear combination calculation.

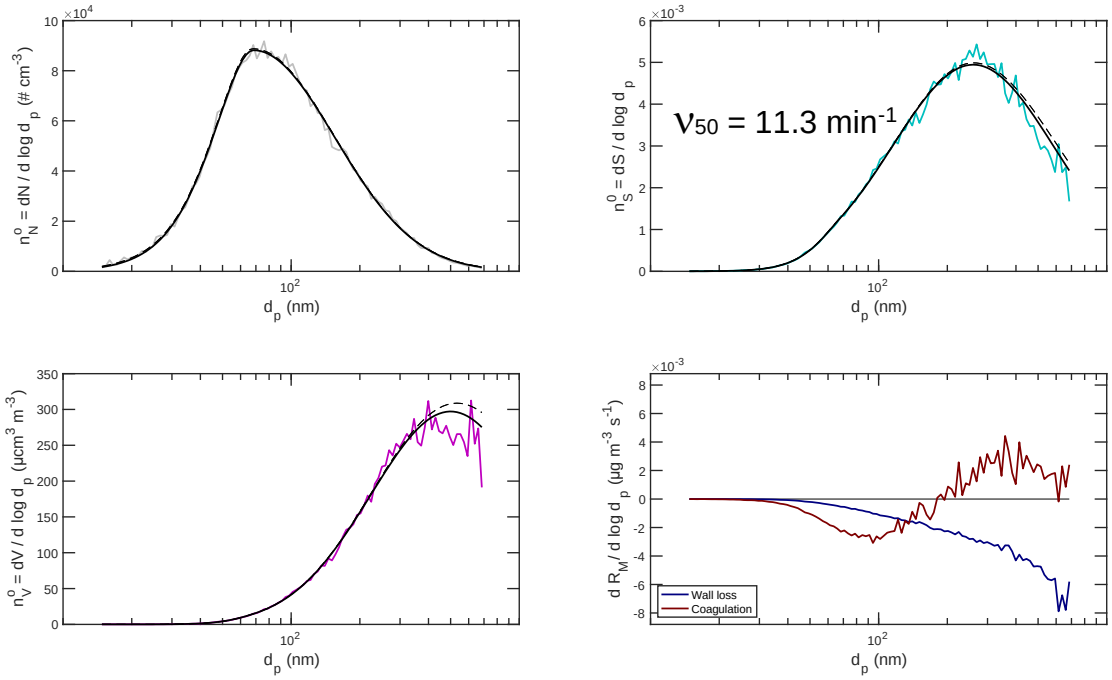

Figure S10: Particle size distributions and gain/loss rates from the mixing experiment conducted with the erythritol/black carbon and ^{13}C-glucose/ammonium sulfate populations at 10 ºC and 90 % RH. These panels represent data collected immediately after introducing the erythritol/black carbon population into the chamber, when the particle concentration of both populations together would be the highest. The top left panel shows a particle number distribution with a size mode just under 100 nm and a total particle concentration of ~45,000 #/cm^3. The top right panel shows a particle surface area concentration with a size mode around 250 nm and particle surface area concentration of 2.2e-3 m^2/m^3. The bottom left panel shows the particle volume distribution with a size mode around ~500 nm and a particle volume concentration of ~170 μcm^3 m^{-3}. The bottom right panel shows the calculated particle gain/loss rates in terms of particle mass for coagulation and wall losses. This experiment had a total coagulation loss rate of ~18,000 # cm^{-3} hr^{-1}, however the resulting change in particle mass in particles that are seen by the AMS (≥100 nm) is ~3.2x10^{-5} μg cm^{-3} hr^{-1}. Thus, coagulation is not a major source of mixing in this system.

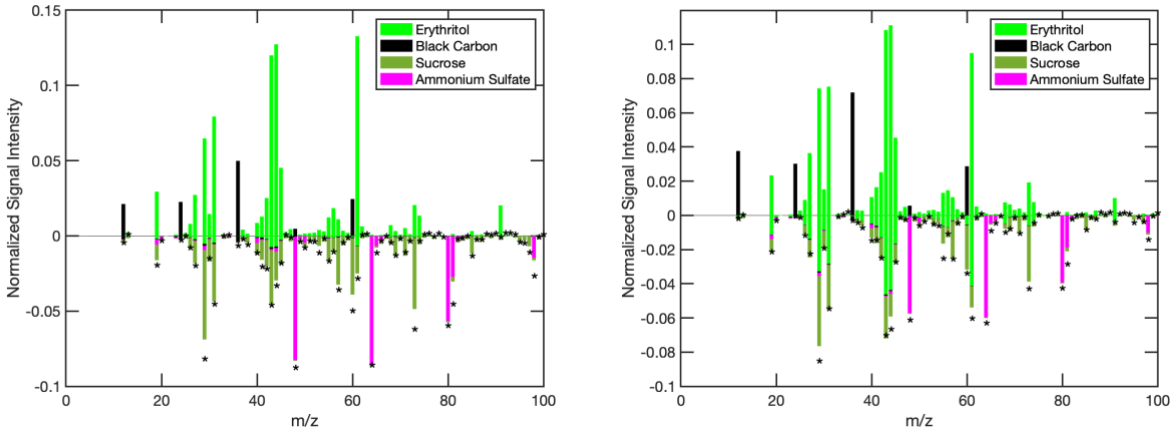

Figure S11: Aggregated mass spectra of particle populations from the start (left) to end (right) of mixing event. We added particles with volatile erythritol (green) coating black carbon (black) into a chamber containing particles with non-volatile sucrose (olive) coating ammonium sulfate (magenta) at 10 ºC and 90 % RH. Over the course of the experiment the erythritol signal decreases relative to the black carbon signal, made evident by the decreases in the green *m/z* sticks relative to the black m/z sticks. There is also an observed increase in corresponding *m/z* sticks in the sucrose/ammonium sulfate population. This suggests that mixing is generally uninhibited for this system at these conditions. The asterisks represent the residual from the linear combination calculation.

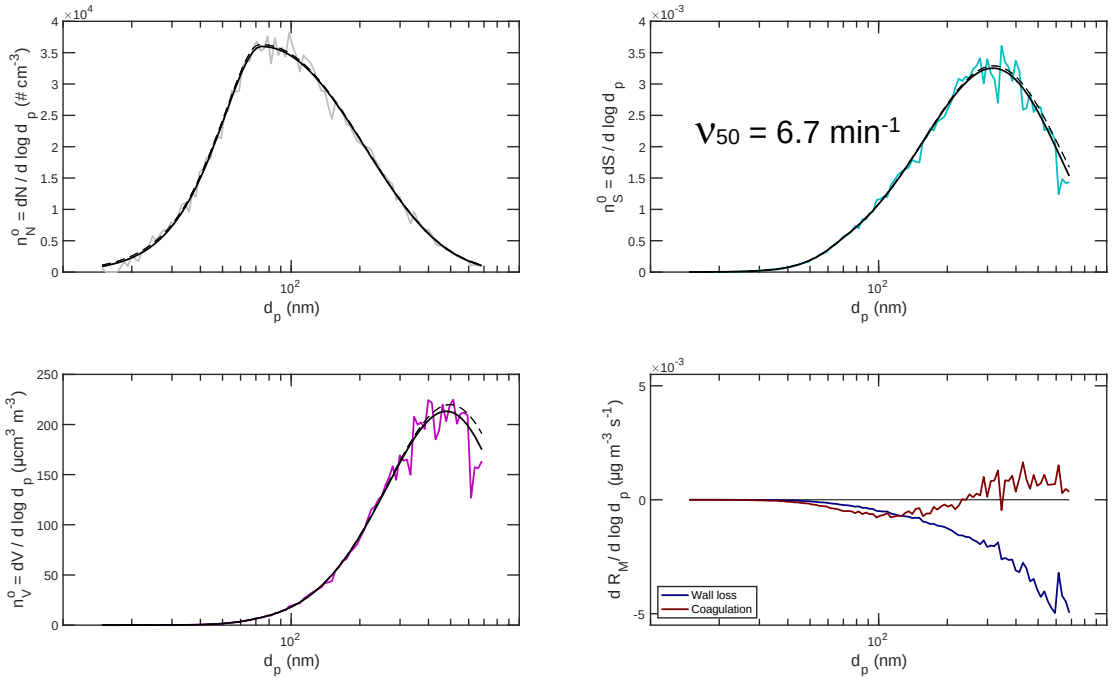

Figure S12: Particle size distributions and gain/loss rates from the mixing experiment conducted with the erythritol/black carbon and sucrose/ammonium sulfate populations at 10 ºC and 90 % RH. These panels represent data collected immediately after introducing the erythritol/black carbon population into the chamber, when the particle concentration of both populations together would be the highest. The top left panel shows a particle number distribution with a size mode just under 100 nm and a total particle concentration of ~30,000 #/cm^3. The top right panel shows a particle surface area concentration with a size mode around 250 nm and particle surface area concentration of 2.2e-3 m^2/m^3. The bottom left panel shows the particle volume distribution with a size mode between 400-500 nm and a particle volume concentration of ~110 µcm^3 m^{-3}. The bottom right panel shows the calculated particle gain/loss rates in terms of particle mass for coagulation and wall losses. This experiment had a total coagulation loss rate of ~3,600 # cm^{-3} hr^{-1}, however the resulting change in particle mass in particles that are seen by the AMS (≥100 nm) is ~1.2x10^{-5} µg cm^{-3} hr^{-1}. Thus, coagulation is not a major source of mixing in this system.

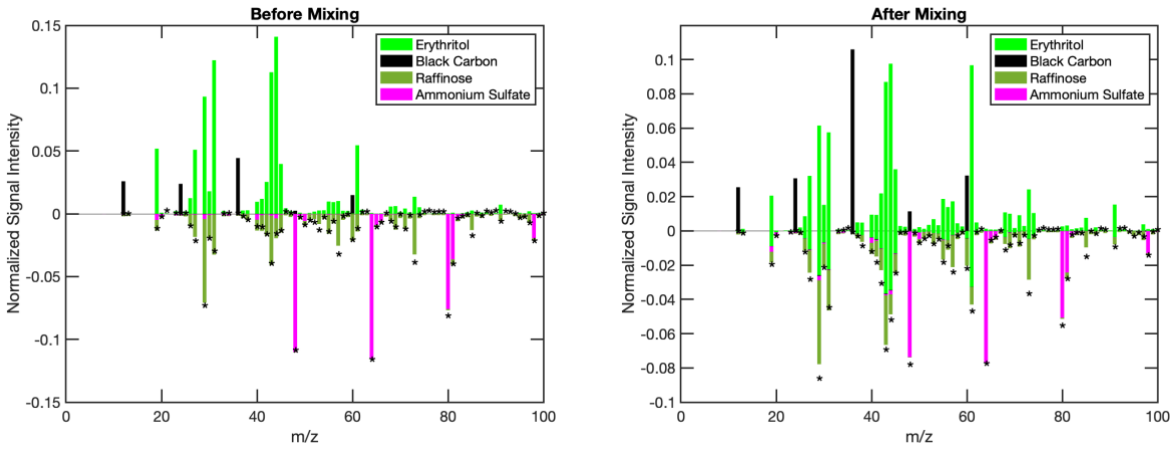

Figure S13: Aggregated mass spectra of particle populations from the start (left) to end (right) of mixing event. We added particles with volatile erythritol (green) coating black carbon (black) into a chamber containing particles with non-volatile raffinose (olive) coating ammonium sulfate (magenta) at 10 ºC and 90 % RH. Over the course of the experiment the erythritol signal decreases relative to the black carbon signal, made evident by the decreases in the green *m/z* sticks relative to the black m/z sticks. There is also an observed increase in corresponding *m/z* sticks in the raffinose/ammonium sulfate population. This suggests that mixing is generally uninhibited for this system at these conditions. The asterisks represent the residual from the linear combination calculation.

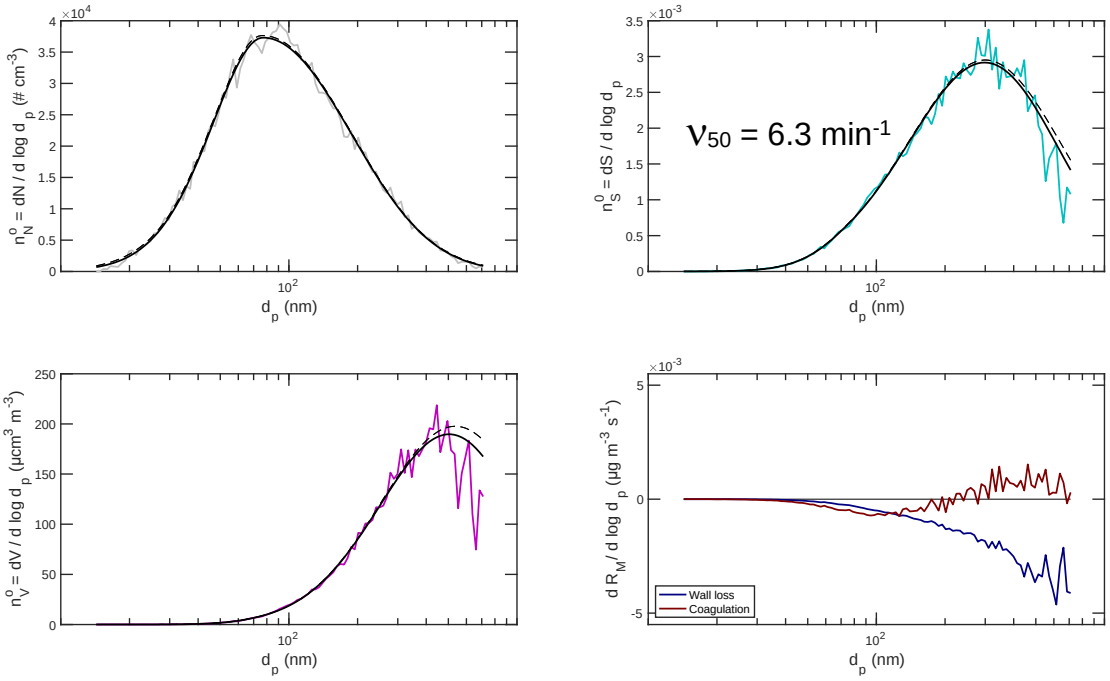

Figure S14: Particle size distributions and gain/loss rates from the mixing experiment conducted with the erythritol/black carbon and raffinose/ammonium sulfate populations at 10 ºC and 90 % RH. These panels represent data collected immediately after introducing the erythritol/black carbon population into the chamber, when the particle concentration of both populations together would be the highest. The top left panel shows a particle number distribution with a size mode just under 100 nm and a total particle concentration of ~30,000 #/cm^3. The top right panel shows a particle surface area concentration with a size mode around 250 nm and particle surface area concentration of 2.1e-3 m^2/m^3. The bottom left panel shows the particle volume distribution with a size mode between 400-500 nm and a particle volume concentration of ~100 µcm^3 m^{-3}. The bottom right panel shows the calculated particle gain/loss rates in terms of particle mass for coagulation and wall losses. This experiment had a total coagulation loss rate of ~3,600 # cm^{-3} hr^{-1}, however the resulting change in particle mass in particles that are seen by the AMS (≥100 nm) is ~7.2x10^{-6} µg cm^{-3} hr^{-1}. Thus, coagulation is not a major source of mixing in this system.