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Table S2. Flux correlation factors for organic aerosols (OOA, HOA and BBOA), inorganic aerosols (nitrate ( $\text{NO}_3^-$ ) and sulfate ( $\text{SO}_4^{2-}$ )) and trace gas ( $\text{CO}_2$ ), measured at SIMAT flux tower site. The best correlation was found to be between the fluxes of nitrate aerosols and BBOA, and HOA and  $\text{CO}_2$ .

	HOA	BBOA	$\text{SO}_4^{2-}$	$\text{NO}_3^-$	$\text{CO}_2$
OOA	0.37	-0.45	-0.07	-0.31	0.43
HOA		-0.47	-0.26	-0.55	<b>0.74</b>
BBOA			0.55	<b>0.80</b>	-0.49
$\text{SO}_4^{2-}$				0.31	-0.35
$\text{NO}_3^-$					-0.33





Figure S1. SIMAT institute building with a 25 m scaffolding tower on the roof.



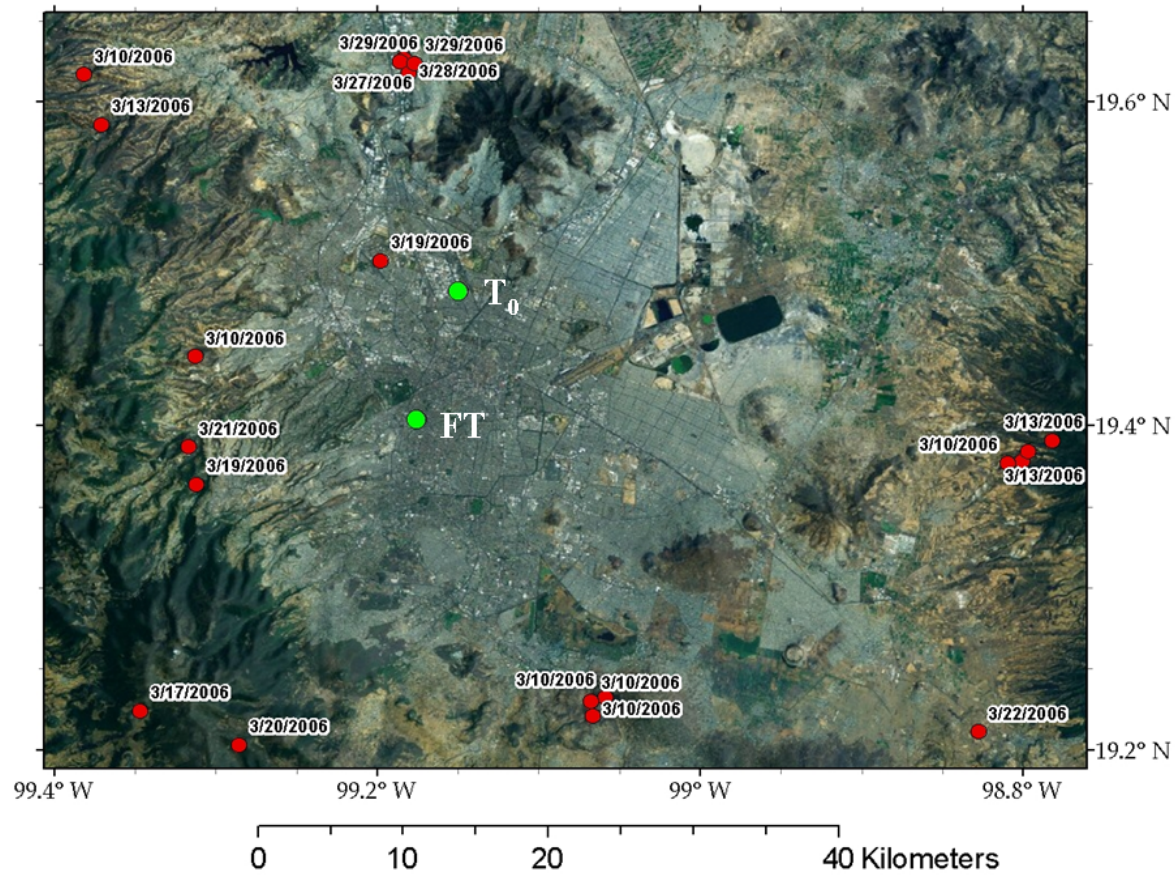


Figure S2. Dated fire incidences (red dots) around and in Mexico City during March 2006. Green dots indicate the measurement sites SIMAT flux tower (FT) and T<sub>0</sub> supersite (T<sub>0</sub>).



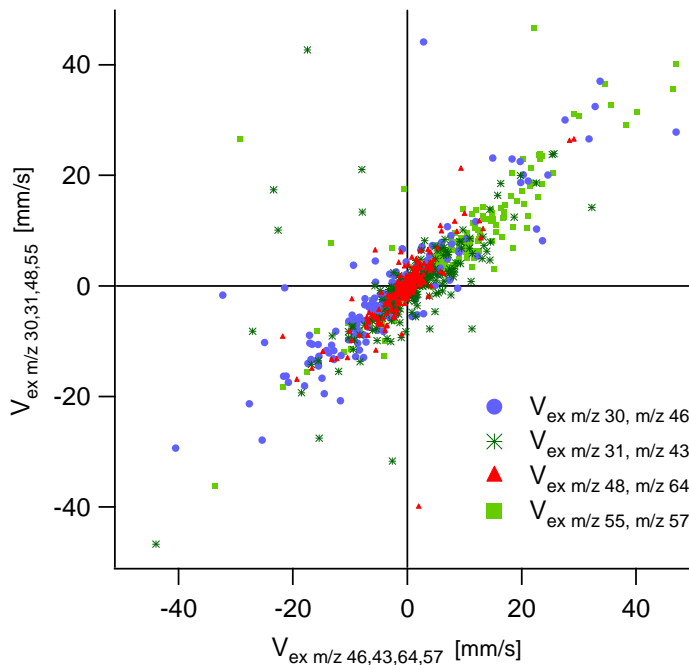


Figure S3. Exchange velocity correlation analysis for measured  $m/z$ 's: nitrate  $m/z$  30 vs.  $m/z$  46 (blue circles), sulfate  $m/z$  48 vs.  $m/z$  64 (red triangles), organic  $m/z$  31 vs.  $m/z$  43 (dark green stars) and  $m/z$  55 vs.  $m/z$  57 (green squares).



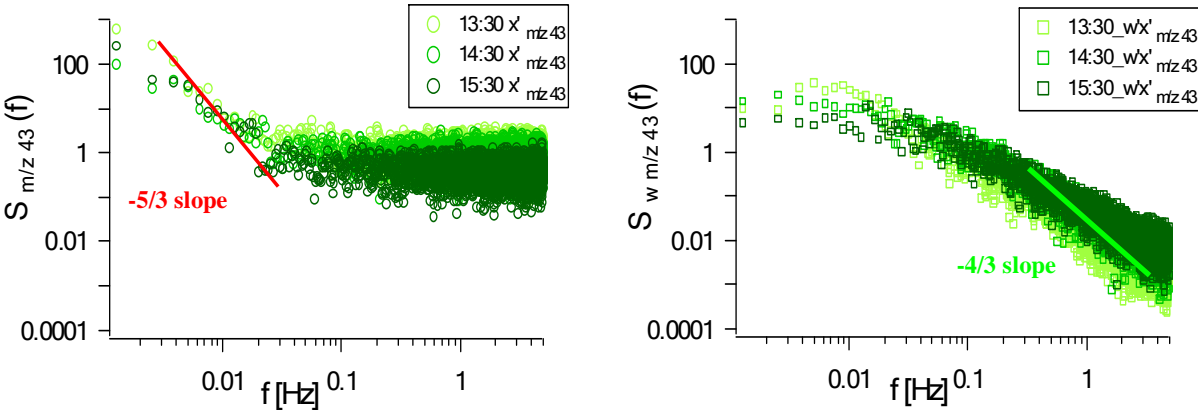


Figure S4. Spectral (left) and cospectral (right) analyses of  $m/z$  43 for three different periods during the daytime: 13:30-14:00, 14:30-15:00 and 15:30-16:00.



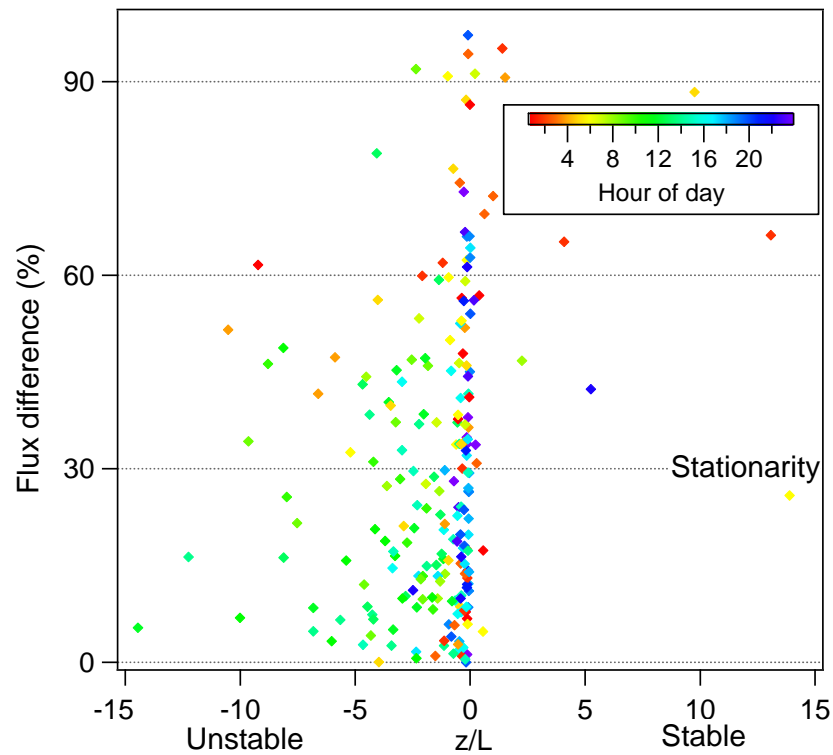
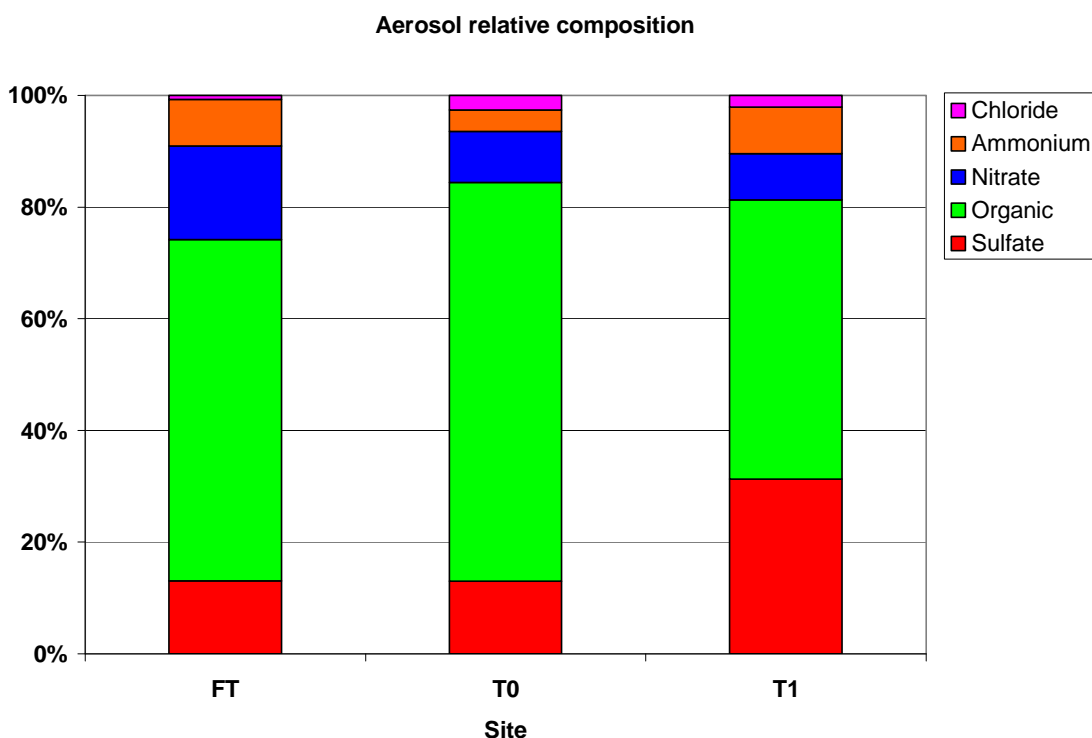


Figure S5. Stationarity test for heat flux. 75% of the periods meet the stationarity criteria. The difference between the 30-minute and  $6 \times 5$ -minute averaged fluxes of 0-30% indicate the threshold where the stationarity criteria was met and exceeded, while the difference of 30-60% means that these periods have an acceptable quality.





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 44 Figure S6. Relative composition of submicron non-refractory aerosol average concentrations at  
 45 the SIMAT flux tower site and T<sub>0</sub> and T<sub>1</sub> supersites: sulfate (red), chloride (pink), ammonium  
 46 (orange), nitrate (blue) and organic (green). Comparing the PM<sub>2.5</sub> concentrations at supersites T<sub>0</sub>  
 47 and T<sub>1</sub> (Querol et al., 2008) and submicron non-refractory aerosols at SIMAT, organics were the  
 48 dominant species out of the five components at all sites. Sulfate was much lower in densely  
 49 populated SIMAT and T<sub>0</sub> sites, but was substantially higher in the further T<sub>1</sub> supersite (30 km  
 50 North from T<sub>0</sub>). Chloride aerosols were on the order of a few percent at all 3 locations. At  
 51 SIMAT we measured significantly higher fraction of nitrate aerosols.  
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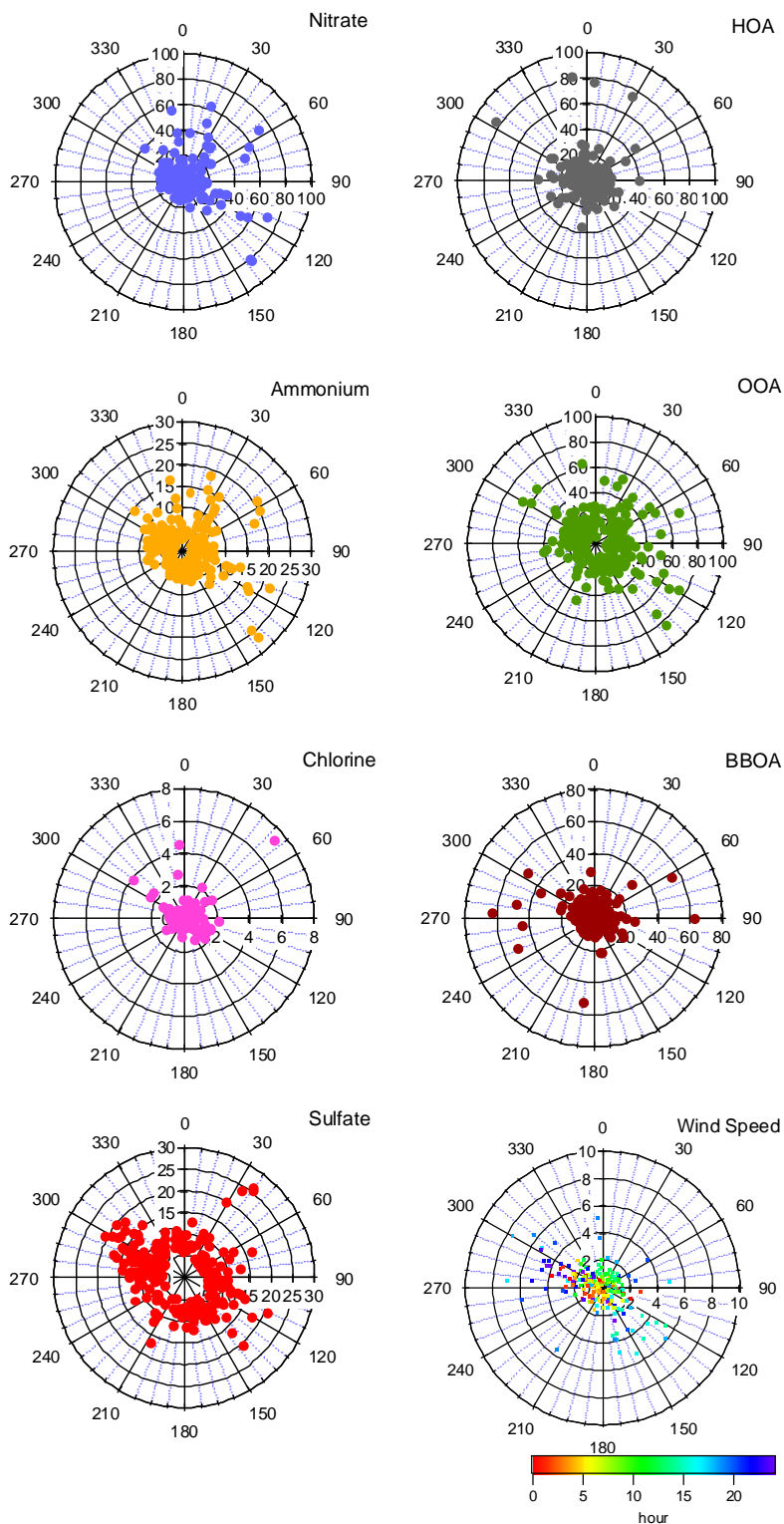


Figure S7. Concentration ( $\mu\text{g m}^{-3}$ ) rose plots of inorganic aerosols (nitrate, chlorine, ammonium and sulfate) and organic aerosols (biomass burning, oxygenated and hydrocarbon-like) and the corresponding wind rose plot of the wind speed ( $\text{m s}^{-1}$ ) distribution. The data at 30 minute averages.



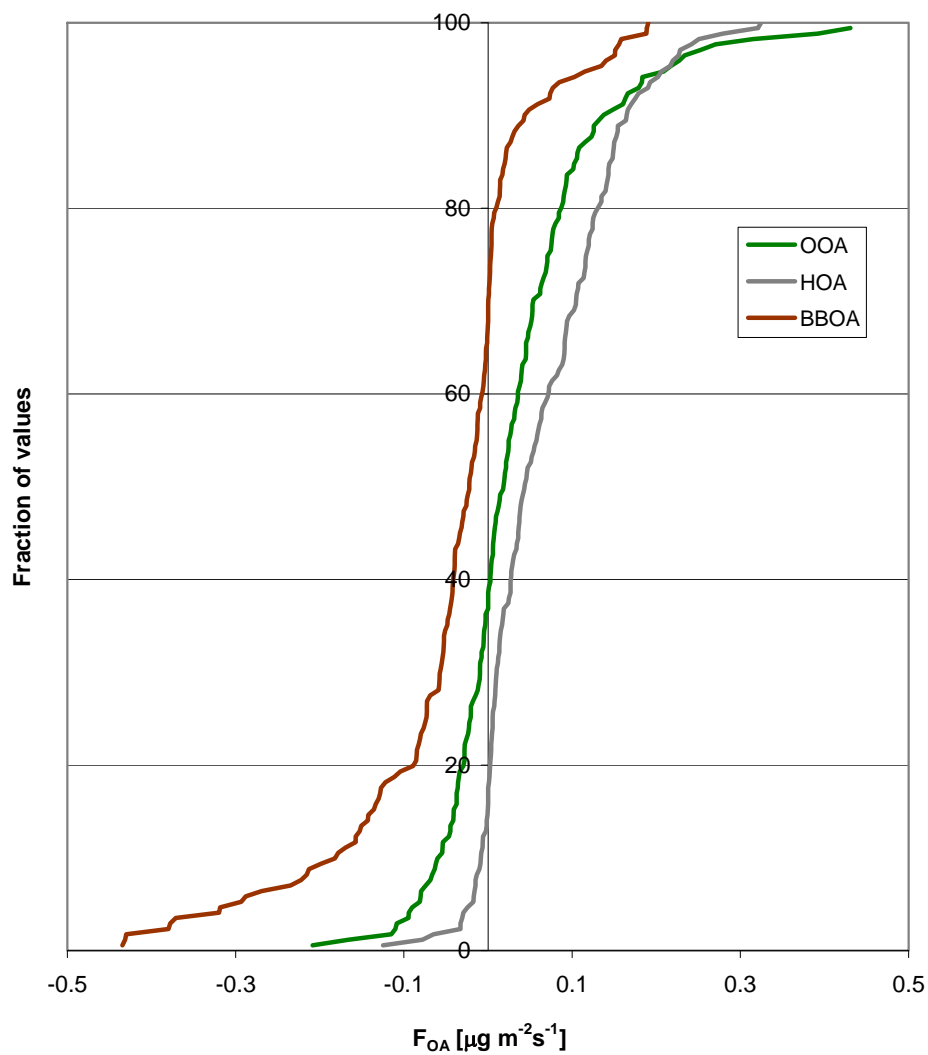


Figure S8. Incidence in percent of organic aerosol component fluxes during the measurement period.



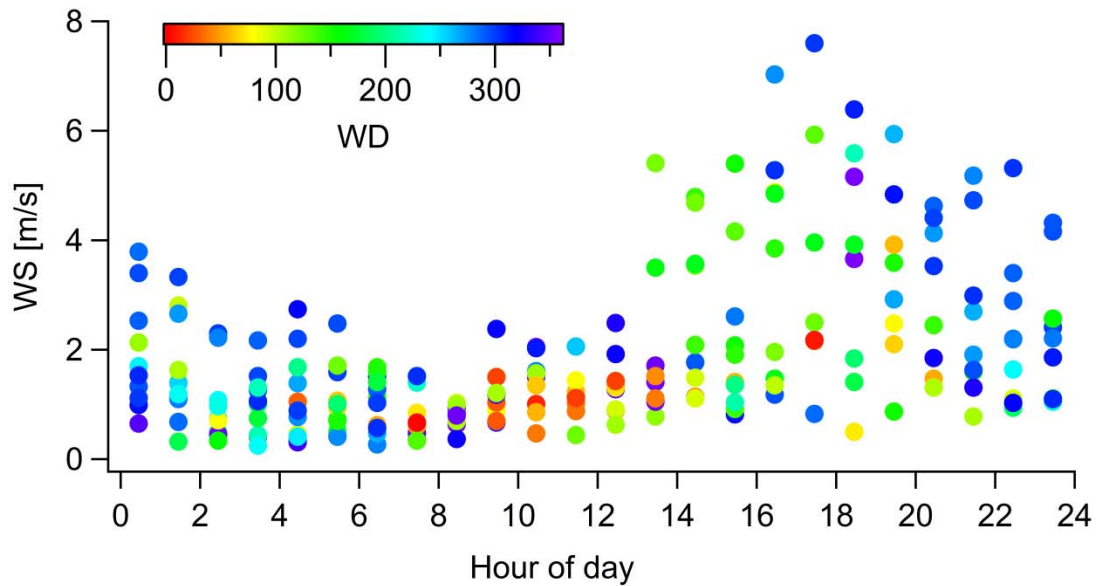


Figure S9. Wind speed (WS) and wind direction (WD) analysis in terms of time of day. During the early morning the wind speed is low and mainly from southwest and northwest directions. In the morning to noon period the wind speed starts increasing and shifts more towards northeast. During the afternoon to late afternoon the winds speeds pick up to 6-8 m/s and blow mainly from south, but there is some eastern flow and high flow from the north. In the evening the wind speed drops and shifts to more northern flow.