Supplement Table 1. Slope, intercept and $R^{2}$ of the regression line for the scattering comparison between the OPC and nephelometer at 450, 550 and 700 nm.

| $\lambda(\mathrm{nm})$ | Slope | Intercept | $\mathrm{R}^{2}$ |  |  |
| :---: | :---: | :---: | :---: | :---: | :---: |
| Central Mexico |  |  |  |  |  |
| 450 |  |  |  |  |  |
| 550 | 1.08 | -1.61 | 0.86 |  |  |
| 700 | 1.11 | -1.14 | 0.85 |  |  |
|  | US West Coast |  |  |  | 0.84 |
|  |  | -1.29 |  |  |  |
| 450 | 0.94 | 0.37 | 0.89 |  |  |
| 550 | 0.96 | 0.16 | 0.86 |  |  |
| 700 | 0.97 | 0.03 | 0.83 |  |  |

The intercept is in $\mathrm{Mm}^{-1}$.

Supplement Table 2. Fit to the aerosol hygroscopicity / chemical composition relationship with $\kappa$ on a logarithmic scale.

| $D_{d}(\mathrm{~nm})$ | $\log _{10} K$ | $K$ at $\mathrm{OMF}=0$ | $K$ at $\mathrm{OMF}=1$ | K variability (\%) |
| :---: | :---: | :---: | :---: | :---: |
| Central Mexico |  |  |  |  |
| 50 | -0.29-0.45*OMF | 0.51 | 0.18 | 25 |
| 100 | -0.43-0.44*OMF | 0.37 | 0.13 | 34 |
| 200 | -0.30-0.57*OMF | 0.50 | 0.13 | 30 |
| 300 | -0.39-0.29*OMF | 0.41 | 0.21 | 31 |
| US West Coast |  |  |  |  |
| 50 | -0.29-0.87*OMF | 0.52 | 0.07 | 49 |
| 100 | -0.29-0.70*OMF | 0.51 | 0.10 | 36 |
| 200 | N/A | N/A | N/A | N/A |
| 300 | -0.30-0.32*OMF | 0.50 | 0.24 | 39 |

[^0]

Supplement Figure 1.
Comparison of the scattering coefficient at 550 nm between the OPC and the nephelometer over 7-170 ${ }^{\circ}$, averaged over 30 seconds, for Central Mexico (left panel) and the US West Coast (right). The dashed lines represent the linear regression: $y=1.11 x-1.14\left(R^{2}=0.85\right)$ and $y=0.96 x+0.16\left(R^{2}=0.86\right)$ for the two regions respectively.

Central Mexico


Supplement Figure 2.
Same as Figure 3c but for the other TDMA dry diameters.



US West Coast


US West Coast


Supplement Figure 3.
Same as Figure 3d but for the other TDMA dry diameters. The 200-nm scans, scarcely made over the US West Coast, are not shown.

Central Mexico


Central Mexico


Central Mexico


Central Mexico


Supplement Figure $4 . \quad$ Same as Supplement Figure 2 but with a logarithmic scale for $\kappa$ and the critical diameter.

US West Coast


Same as
Supplement Figure 5.
Supplement Figure 3 but with a logarithmic scale for $\kappa$ and the critical diameter.

US West Coast




[^0]:    $D_{d}$ is the dry diameters selected with the TDMA, OMF is the organic mass fraction of non-refractory component of submicron aerosols, $\kappa$ is the hygroscopicity parameter. $\kappa$ variability is the apparent relative variability of individual data points from the fit, ( $\left.10^{\text {RMSlog }}-1\right)^{*} 100$, where RMSlog is the root mean square of the differences in $\log _{10} K$ between the fit and the individual data. See Figure S4 for the fit lines.

