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Characteristics and sources of aerosol aminiums over the eastern coast of China: insights from the integrated observations in acoastal city, adjacent island and surrounding marginal seas

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Total suspended particle (TSP) sample collection and measurement

TSP samples were collected at Huaniao Island during the autumn of 2016 (24 Oct. – 1 Dec.) and the summer of 2017 (21 Jun. – 9 Jul. and 28 Aug. – 12 Sep.) by a high-volume sampler (1050 L min^{-1}, HY-1000D, Hengyuan) using cellulose filters. The sampling duration was commonly 24 hours. 1/32 of TSP sample filter was cut and extracted ultrasonically by 20 mL of ultrapure water (18.25 MΩ cm^{-1}). The extract was then filtered and analyzed for anions using Ion Chromatograph.

Calculation of the retention percentage of air mass over the land (R_L)

The three-day backward trajectories starting from Huaniao Island were calculated every 3 hours for each TSP sample (Figure S9). There are a total of 73 endpoints tracking 0 to 72 hours along each trajectory, and then the proportion of endpoints over the land was calculated. In addition, the regions corresponding to longer backward tracking time had weaker influence on the receptor site than the nearby regions, so a weighting factor associated with the backward tracking time was also included. The calculation is shown as below.

$$R_L = \frac{\sum_{i=1}^{N_{land}} e^{-\frac{t_i}{72}}}{\sum_{i=1}^{N_{total}} e^{-\frac{t_i}{72}}} \times 100\%$$

where N_{total} is the total number of trajectory endpoints corresponding to a TSP sample. N_{land} is the total number of trajectory endpoints located over the land. t_i is the backward tracking time with the unit of hour and $e^{-\frac{t_i}{72}}$ is the weighting factor.

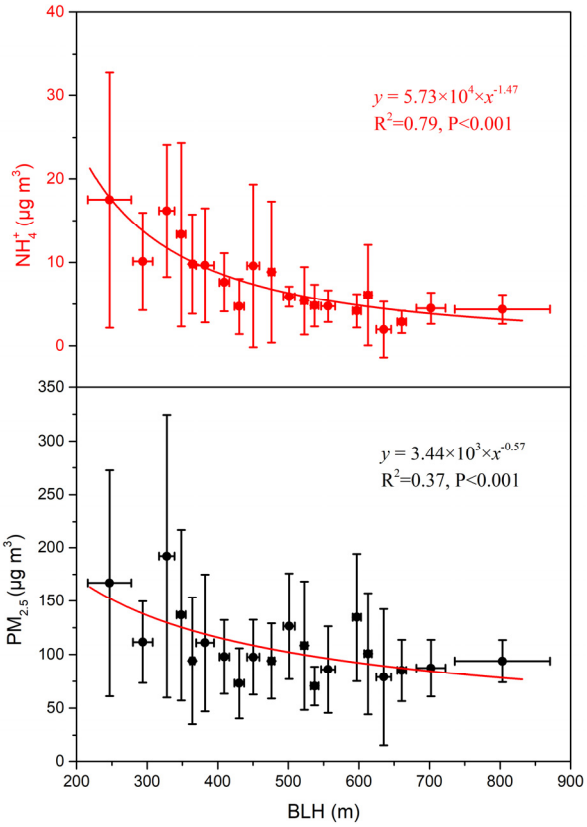

Figure S1. Relationships between concentrations of $PM_{2.5}$ and NH_4^+ and boundary layer height (BLH).

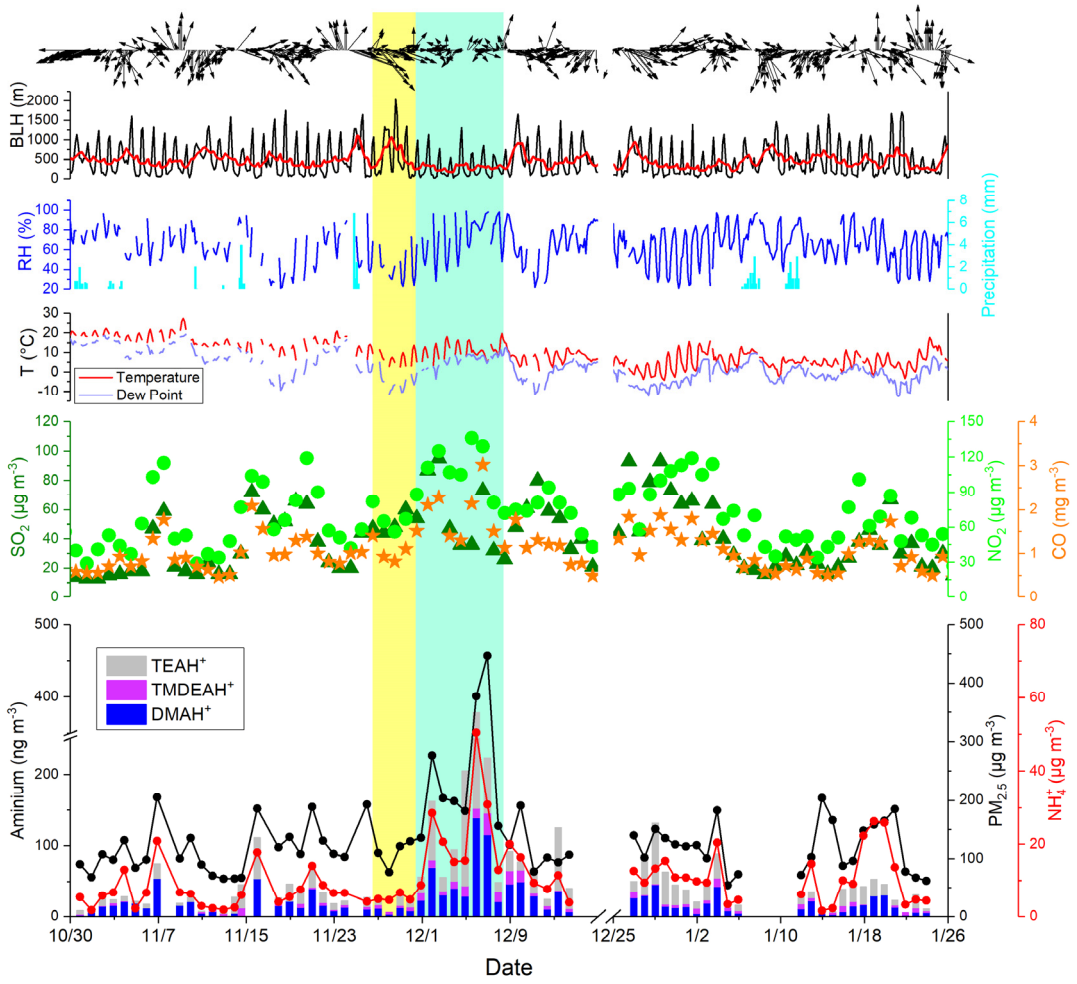

Figure S2. Time series of concentrations of atmospheric species and meteorological factors during the autumn and winter of 2013 in Shanghai.

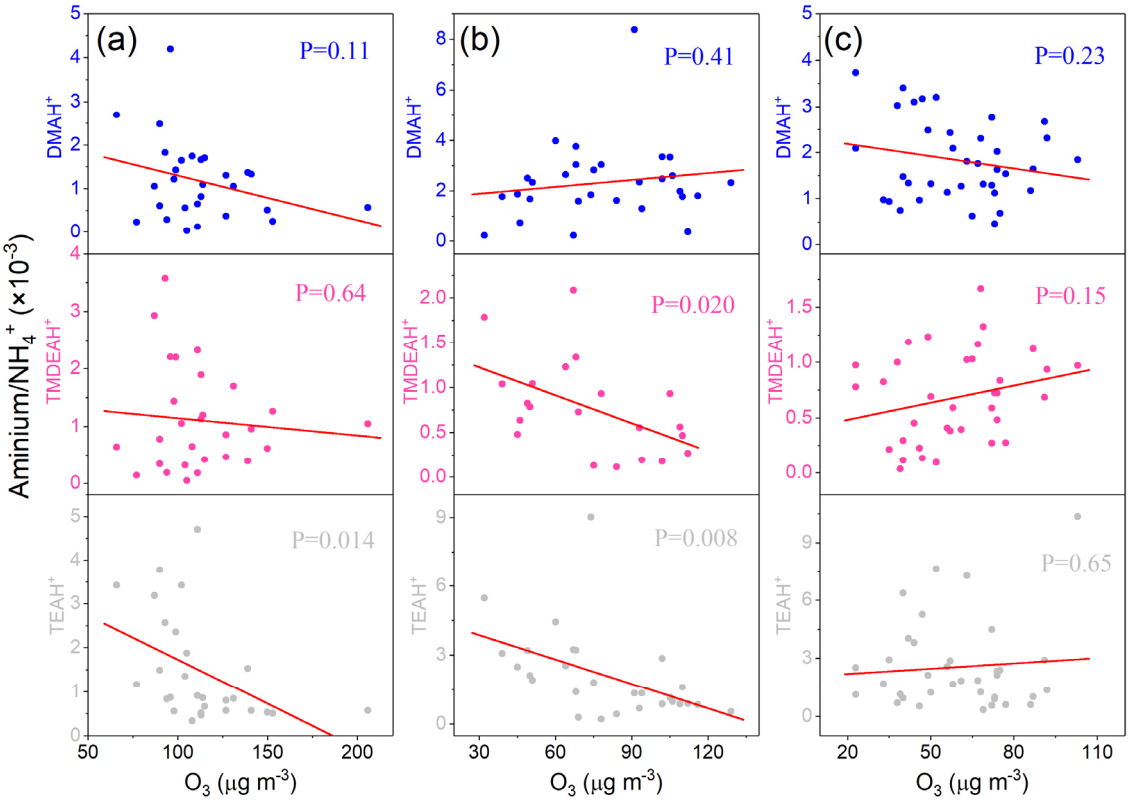

Figure S3. Relationships between mass ratios of aminiums to NH_4^+ and O_3 concentrations over Shanghai in the **(a)** spring, **(b)** autumn and **(c)** winter of 2013.

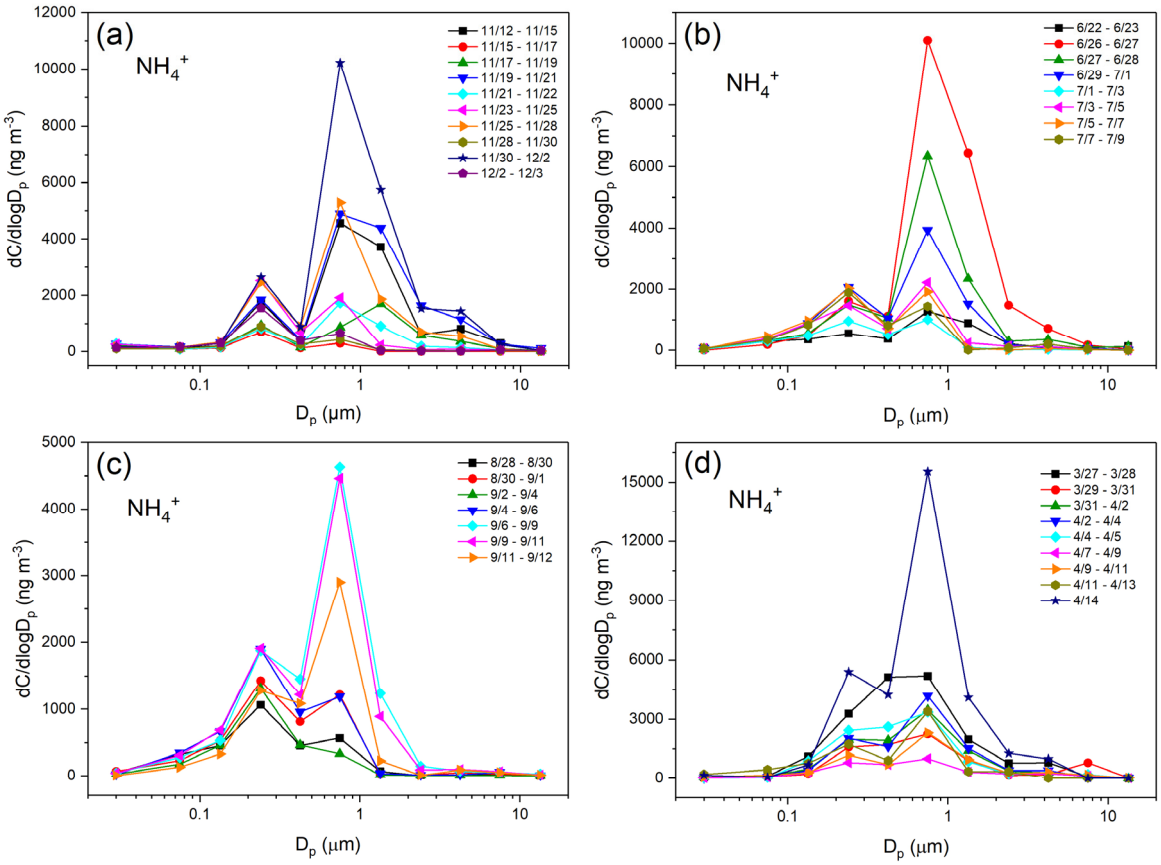

Figure S4. Size distributions of NH_4^+ during different campaigns. **(a):** in the autumn of 2016 at Huaniao Island, **(b):** in early summer of 2017 at Huaniao Island, **(c):** in late summer of 2017 at Huaniao Island, **(d):** in 2017 spring cruise over the Yellow and East China seas.

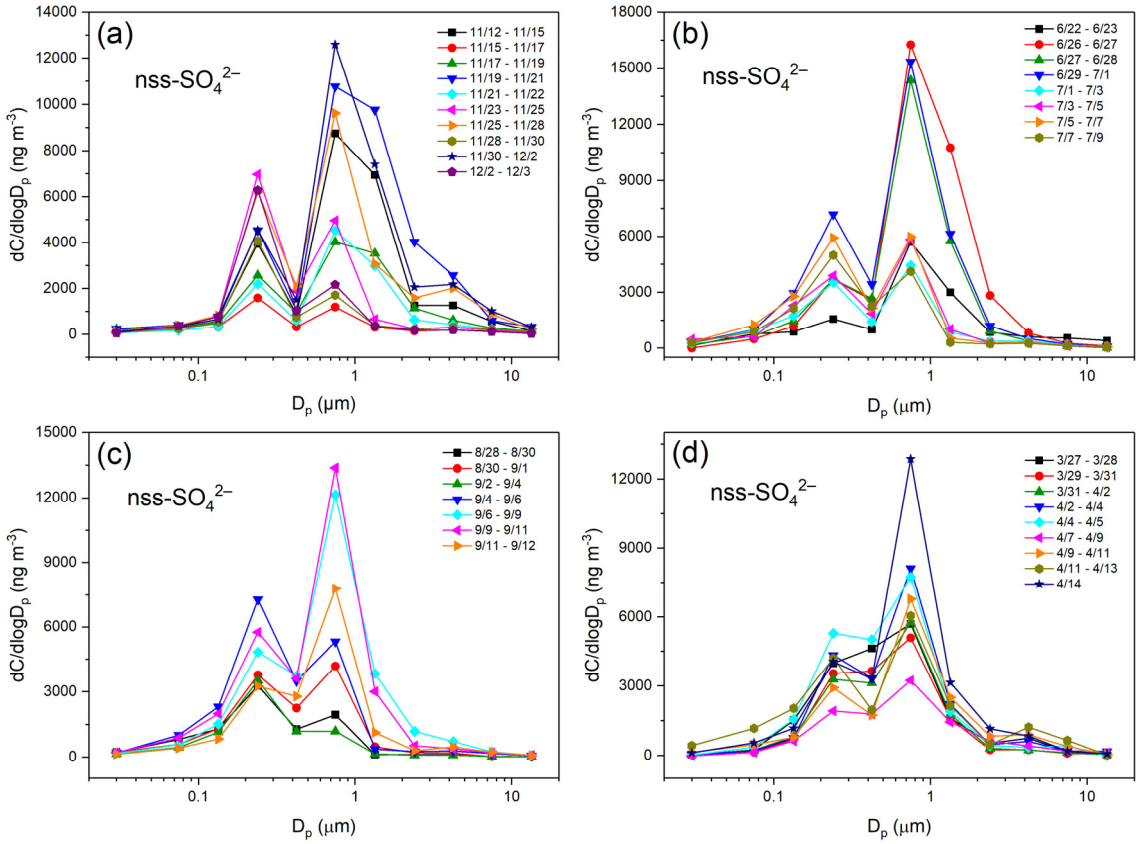

Figure S5. Size distributions of nss-SO_4^{2-} during different campaigns. **(a):** in the autumn of 2016 at Huaniao Island, **(b):** in early summer of 2017 at Huaniao Island, **(c):** in late summer of 2017 at Huaniao Island, **(d):** in 2017 spring cruise over the Yellow and East China seas.

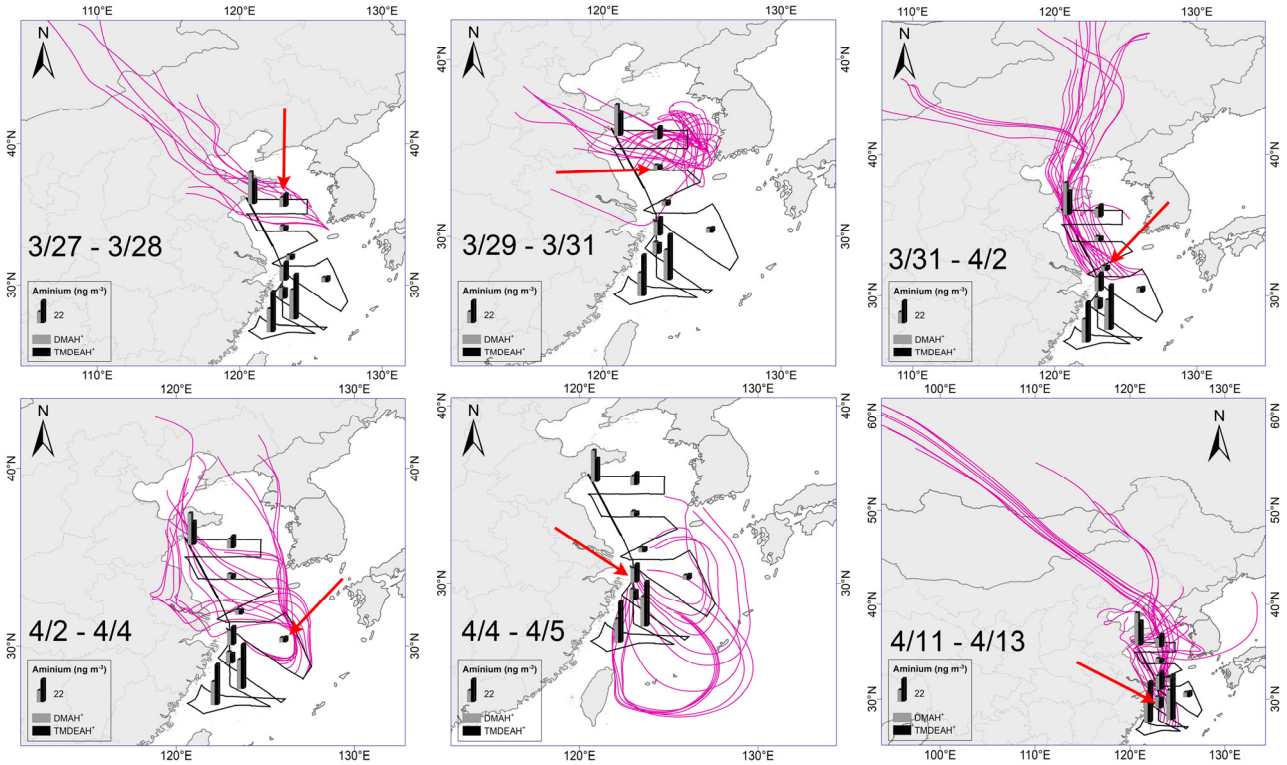

Figure S6. 72-hour air mass backward trajectories for the samples other than those were plotted in Figure 9.

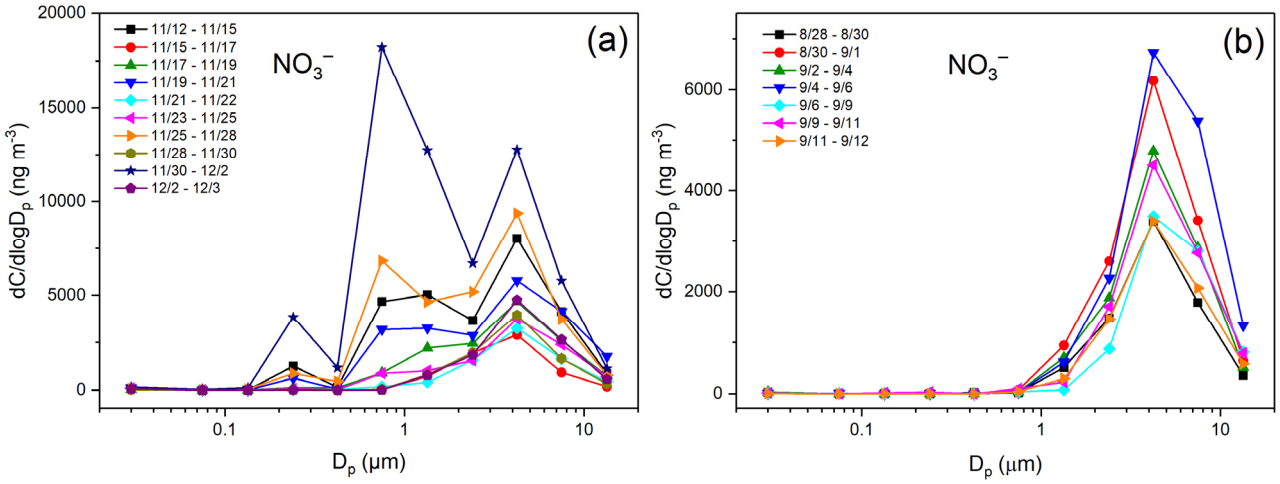

Figure S7. (a) Size distributions of NO_3^- over Huaniao Island in the autumn of 2016. **(b)** Size distributions of NO_3^- over Huaniao Island in the late summer of 2017.

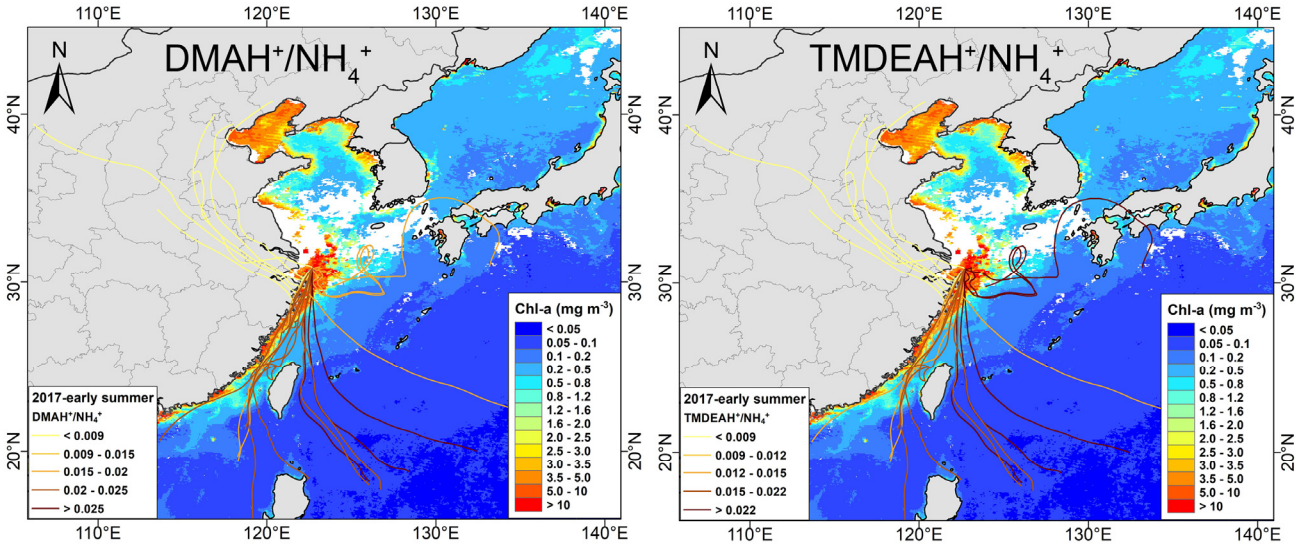

Figure S8. The backward trajectories starting from Huaniao Island during early summer in 2017 and the mass ratio of aminium to NH_4^+ for the corresponding sample.

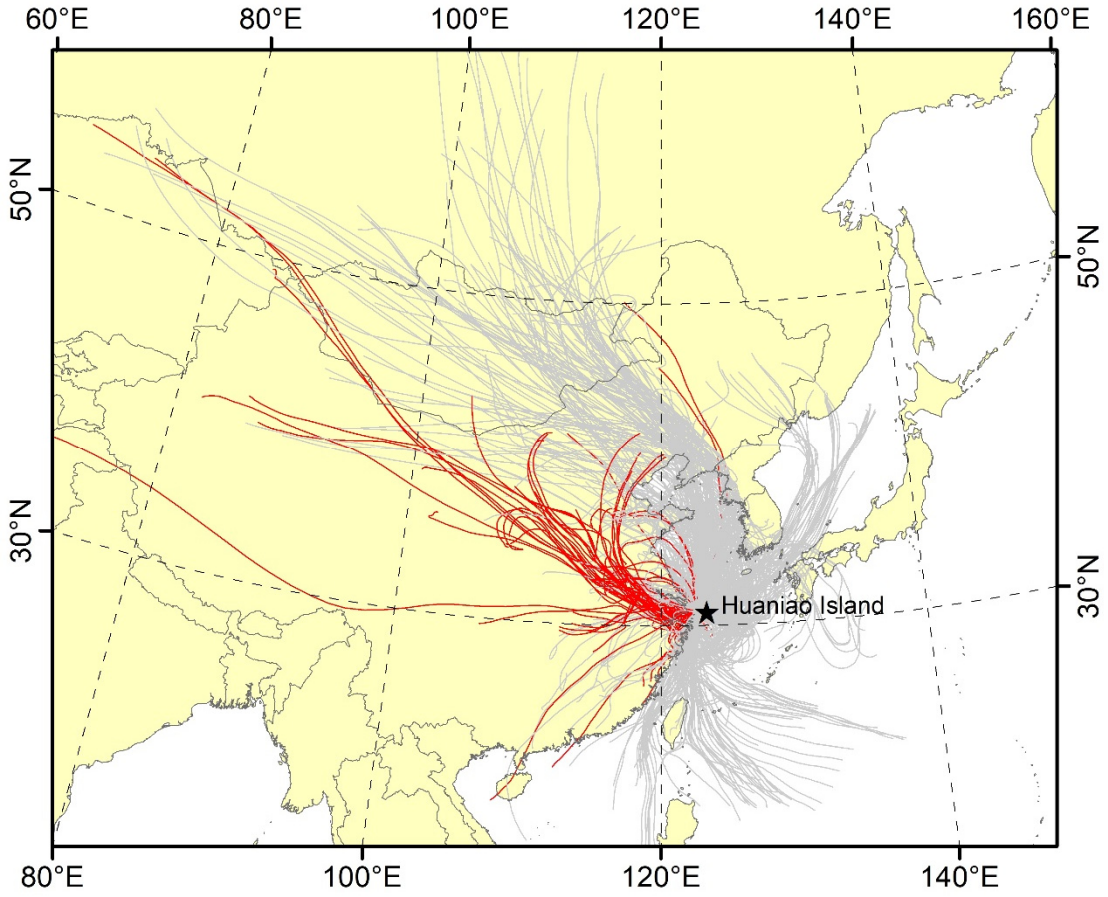

Figure S9. The three-day backward trajectories starting from Huaniao Island during the autumn of 2016 and the summer of 2017 corresponding to TSP samples. A new trajectory was calculated every 3 hours during the sampling period of each sample. The red trajectories corresponded to the samples identified as terrestrial dominated.