*Supplement of*

# Persistent primary organic tar particles during the regional wintertime hazes in North China: insights into their aging and optical changes

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Table S1. Detailed information on individual particle samples collected at the Gucheng rural and Beijing urban sites.

| Location | Date | Local time | Sampling duration (s) | Temperature (°C) | RH (%) | Pressure (hPa) | WS (m s$^{-1}$) | Pollution level |
|---|---|---|---|---|---|---|---|---|
| Gucheng | 21 Nov. 2016 | 12:00 | 180 | −0.6 | 100 | 1018 | 4 | Clean |
| | 22 Nov. 2016 | 8:06 | 180 | −4.7 | 97 | 1012 | 0.9 | Clean |
| | 22 Nov. 2016 | 19:22 | 120 | −3.8 | 47 | 1036 | 0.6 | Heavy (Early stage) |
| | 23 Nov. 2016 | 10:50 | 15 | 0.9 | 51 | 1034 | 0.6 | Heavy (Early stage) |
| | 24 Nov. 2016 | 8:06 | 15 | −6.4 | 94 | 1028 | 0 | Heavy (Middle stage) |
| | 25 Nov. 2016 | 3:00 | 15 | −2.0 | 79 | 1022 | 1.6 | Heavy (Middle stage) |
| | 25 Nov. 2016 | 8:07 | 10 | −4.7 | 93 | 1023 | 0 | Heavy (Middle stage) |
| | 25 Nov. 2016 | 15:19 | 10 | 1.4 | 77 | 1022 | 1.6 | Heavy (Late stage) |
| | 25 Nov. 2016 | 20:44 | 10 | 1.1 | 84 | 1022 | 1.7 | Heavy (Late stage) |
| | 26 Nov. 2016 | 3:00 | 8 | −1.4 | 95 | 1018 | 0 | Heavy (Late stage) |
| | 26 Nov. 2016 | 8:15 | 8 | −1.9 | 98 | 1018 | 0 | Heavy (Late stage) |
| | 26 Nov. 2016 | 12:27 | 10 | 5.5 | 70 | 1017 | 0.8 | Heavy (Late stage) |
| Beijing | 22 Nov. 2016 | 19:00 | 60 | −1.8 | 33 | 1039 | 0.5 | Clean |
| | 24 Nov. 2016 | 19:00 | 15 | 2.9 | 41 | 1026 | 0.9 | Moderate |
| | 25 Nov. 2016 | 12:00 | 20 | 3.8 | 42 | 1026 | 1.5 | Moderate |
| | 25 Nov. 2016 | 22:00 | 10 | 3.1 | 72 | 1024 | 0 | Heavy |
| | 26 Nov. 2016 | 9:00 | 10 | 2.4 | 75 | 1022 | 0 | Heavy |
| | 26 Nov. 2016 | 16:00 | 10 | 7 | 45 | 1019 | 0 | Heavy |
| | 26 Nov. 2016 | 22:00 | 10 | 3.8 | 71 | 1020 | 0 | Heavy |

Table S2. Summary of the average meteorological parameters and mass concentrations of PM$_{2.5}$, aerosol chemical species, organic aerosol (OA) factors, and gaseous pollutants in different periods at the Gucheng rural and Beijing urban sites.

| | **Gucheng rural site** | | | | **Beijing urban site** | | |
|---|---|---|---|---|---|---|---|
| | Clean | Heavily polluted | | | Clean | Moderately polluted | Heavily polluted |
| | | Early stage | Middle stage | Late stage | | | |
| *Meteorological parameters* | | | | | | | |
| $T$ (°C) | −1.5 | −2.9 | −1.6 | 0.53 | −2.0 | 2.1 | 3.9 |
| RH (%) | 97 | 63 | 65 | 78 | 58 | 48 | 64 |
| WS (m s$^{-1}$) | 2.8 | 0.51 | 0.53 | 0.50 | 1.4 | 0.38 | 0.28 |
| $P$ (hPa) | 1013.5 | 1032.6 | 1026.9 | 1019.6 | 1029.1 | 1026.5 | 1021.5 |
| *PM$_{2.5}$ and chemical components ($\mu g\ m^{-3}$)[a]* | | | | | | | |
| PM$_{2.5}$ | 39.8 | 288.3 | 312.3 | 396.8 | 10.8 | 111.0 | 281.0 |
| OM | 19.8 | 185.1 | 175.4 | 217.2 | 5.7 | 44.4 | 82.8 |
| SO$_4^{2-}$ | 2.0 | 13.6 | 17.3 | 41.2 | 2.0 | 13.9 | 51.3 |
| NO$_3^-$ | 2.4 | 11.3 | 14.7 | 30.8 | 1.9 | 19.7 | 41.0 |
| NH$_4^+$ | 1.4 | 11.5 | 13.9 | 28.2 | 1.1 | 9.8 | 24.2 |
| Cl$^-$ | 1.2 | 11.1 | 9.8 | 15.9 | 0.46 | 7.2 | 14.7 |
| EC | 1.4 | 8.9 | 8.7 | 10.9 | / | / | / |
| K$^+$ | 0.36 | 1.6 | 1.5 | 2.7 | / | / | / |
| Na$^+$ | 0.63 | 0.41 | 0.42 | 0.92 | / | / | / |
| Mg$^{2+}$ | 0.08 | 0.18 | 0.15 | 0.28 | / | / | / |
| Ca$^{2+}$ | 0.75 | 1.1 | 0.84 | 1.8 | / | / | / |
| *OA factors ($\mu g\ m^{-3}$)[b]* | | | | | | | |
| FFOA | / | / | / | / | 1.0 | 9.2 | 15.6 |
| COA | / | / | / | / | 1.1 | 6.5 | 9.0 |
| BBOA | / | / | / | / | 0.20 | 5.4 | 15.0 |
| OOA | / | / | / | / | 0.74 | 12.7 | 24.1 |
| OPOA | / | / | / | / | 1.9 | 7.7 | 12.2 |
| aqOOA | / | / | / | / | 0.24 | 3.0 | 10.7 |
| *Gaseous pollutants* | | | | | | | |
| CO (mg m$^{-3}$) | 0.44 | 2.4 | 2.9 | 4.3 | 0.54 | 2.8 | 4.1 |
| SO$_2$ ($\mu g\ m^{-3}$) | 10.3 | 48.2 | 55.5 | 55.9 | 6.0 | 23.2 | 21.8 |
| NO$_2$ ($\mu g\ m^{-3}$) | 25.3 | 62.7 | 80.0 | 94.5 | 29.0 | 94.1 | 130.8 |
| O$_3$ ($\mu g\ m^{-3}$) | 36.1 | 10.7 | 10.2 | 10.5 | 42.6 | 11.0 | 7.6 |

[a]The aerosol chemical components at the Gucheng rural site were obtained by offline analysis of PM$_{2.5}$ filter samples and those at the Beijing urban site were obtained by online analysis of NR-PM$_1$ using a high-resolution aerosol mass spectrometer (HR-AMS).

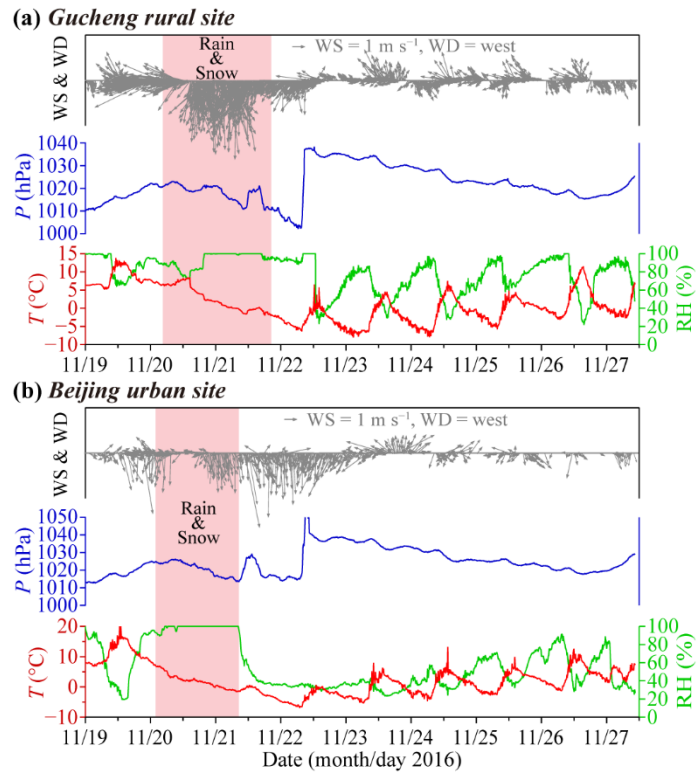[b]OA factors were derived by PMF analysis of HR-AMS data at the Beijing urban site.

Figure S1. Time series of meteorological parameters including temperature (*T*), pressure (*P*), relative humidity (RH), wind speed (WS), and wind direction (WD) at the (a) Gucheng rural site and (b) Beijing urban site.
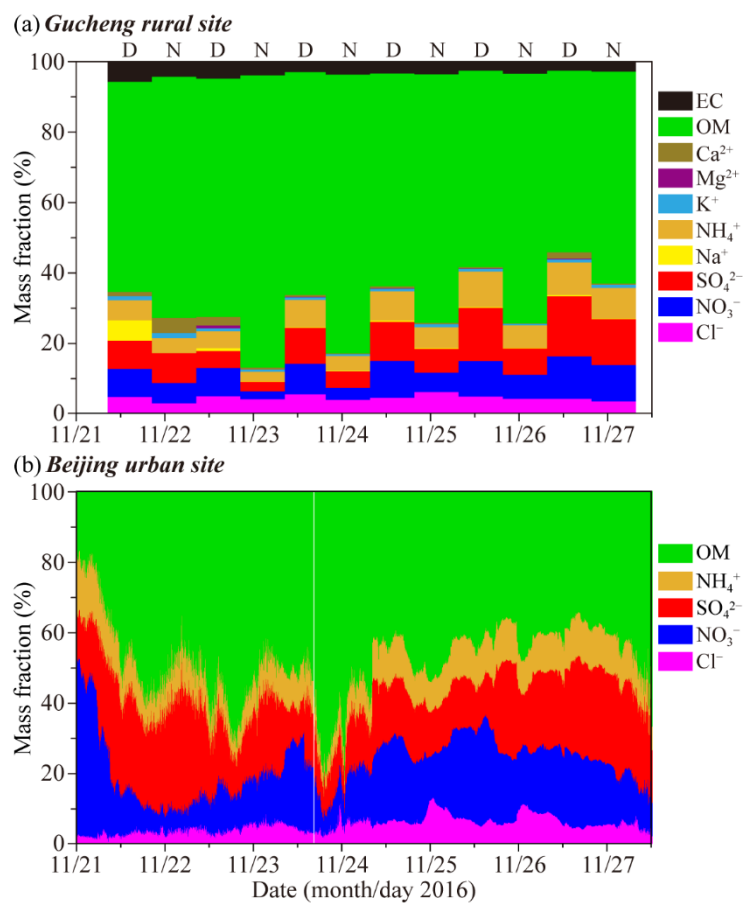
Figure S2. Relative mass fractions of major chemical species in PM$_{2.5}$ at the Gucheng rural site (a) and in NR-PM$_1$ at the Beijing urban site (b).
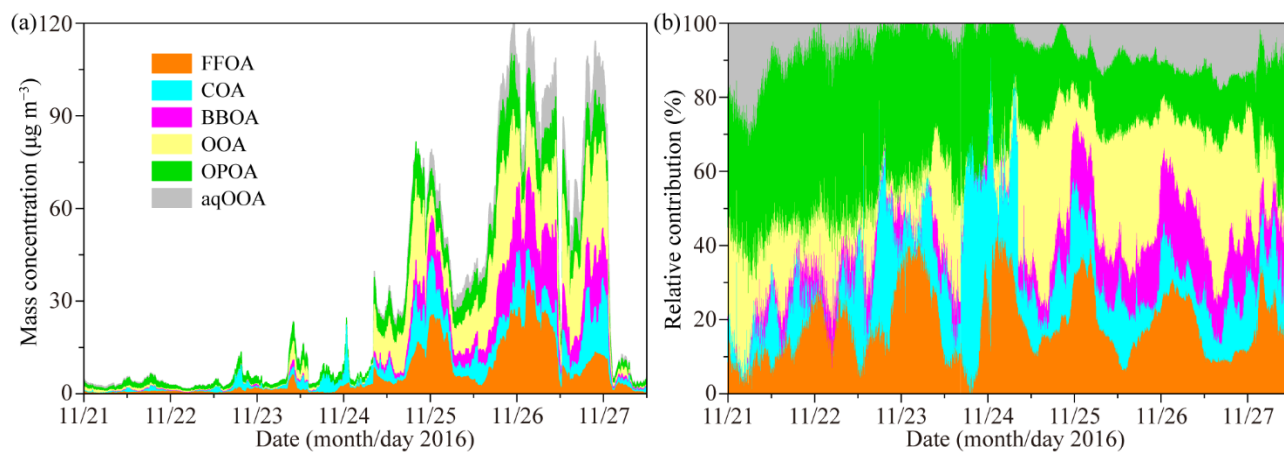
Figure S3. Absolute (a) and relative (b) contributions to organic aerosol (OA) in NR-PM$_1$ from different factors (i.e., FFOA–fossil fuel-related OA, COA–cooking OA, BBOA–biomass burning OA, OOA–oxygenated OA, OPOA–oxidized POA, and aqOOA–aqueous-phase OOA) at the Beijing urban site.