## Supplemental Material

**Table S1**

Field measurements at the eight sites

| Observables | Technique | Max Resolution | Detection Limit |
|---|---|---|---|
| • Gases: | | | ppb |
| NO | CL | 1–min | 0.05 |
| $NO_2$ | Photolysis/CL | 1–min | 0.1 |
| $HNO_3$ | Denuder/Mo reduction/CL | 1–min | 0.1 |
| $NO_y$ | Mo reduction/CL | 1–min | 0.1 |
| $SO_2$ | UV–fluorescence | 1–min | 0.2 |
| $NH_3$ | Denuder/Pt oxidation/CL | 5–min | 0.2 |
| • $iPM_{2.5}$ chemical compositions: | | | $\mu g\ m^{-3}$ |
| $SO_4^{2-}$ | Fe reduction/UV–fluorescence | 5–min | 0.4 |
| $NO_3^-$ | Filter/Mo reduction/CL | 5–min | 0.2 |
| $NH_4^+$ | Filter/Pt oxidation/CL | 5–min | 0.1 |
| • Meteorological conditions: | | | |
| T/RH/SR/BP | Various | 1–min | N/A |
| WS/WD/Precipitation | Various | 1–min | N/A |

CL: chemiluminescence; SR: solar radiation; BP: barometric pressure; WS: wind speed; WD: wind direction; N/A: not applicable.

**Table S2**

The statistics of different precursor gases of $iPM_{2.5}$ by season at the YRK site in 2008-2011

| Season | | $Na^+$ | $TH_2SO_4$ | $TNH_3$ | $THNO_3$ | THCl | $Ca^{2+}$ | $K^+$ | $Mg^{2+}$ | RH | T (K) |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Winter | min | 0.00 | 0.63 | 0.83 | 0.47 | 0.02 | 0.00 | 0.01 | 0.00 | 0.37 | 264.93 |
| | median | 0.02 | 2.19 | 1.84 | 1.99 | 0.04 | 0.02 | 0.02 | 0.00 | 0.61 | 277.18 |
| | max | 0.28 | 5.12 | 5.00 | 8.01 | 0.21 | 0.15 | 0.11 | 0.03 | 0.91 | 290.62 |
| | mean | 0.05 | 2.22 | 2.15 | 2.49 | 0.05 | 0.03 | 0.03 | 0.01 | 0.62 | 277.61 |
| | SD | 0.06 | 1.01 | 1.05 | 1.50 | 0.05 | 0.03 | 0.03 | 0.01 | 0.14 | 5.70 |
| | N | 30 | 30 | 30 | 30 | 30 | 30 | 30 | 30 | 30 | 30 |
| Spring | min | 0.01 | 0.80 | 0.87 | 0.23 | 0.01 | 0.01 | 0.01 | 0.00 | 0.35 | 272.68 |
| | median | 0.03 | 3.36 | 2.71 | 1.62 | 0.04 | 0.03 | 0.03 | 0.01 | 0.64 | 291.29 |
| | max | 0.47 | 12.09 | 9.26 | 3.86 | 0.18 | 0.26 | 0.11 | 0.03 | 0.93 | 299.32 |
| | mean | 0.07 | 3.42 | 2.91 | 1.68 | 0.04 | 0.06 | 0.04 | 0.01 | 0.64 | 289.45 |
| | SD | 0.09 | 2.04 | 1.52 | 0.70 | 0.04 | 0.07 | 0.02 | 0.01 | 0.14 | 6.12 |
| | N | 34 | 34 | 34 | 34 | 34 | 34 | 34 | 34 | 34 | 34 |
| Summer | min | 0.00 | 1.57 | 1.06 | 0.35 | 0.01 | 0.01 | 0.00 | 0.00 | 0.47 | 294.27 |
| | median | 0.02 | 3.98 | 2.87 | 1.34 | 0.03 | 0.03 | 0.02 | 0.01 | 0.71 | 298.60 |
| | max | 0.21 | 13.66 | 7.81 | 3.82 | 0.13 | 0.06 | 0.35 | 0.07 | 0.96 | 301.90 |
| | mean | 0.04 | 4.51 | 3.07 | 1.51 | 0.03 | 0.03 | 0.03 | 0.01 | 0.70 | 298.42 |
| | SD | 0.04 | 2.60 | 1.27 | 0.64 | 0.02 | 0.01 | 0.05 | 0.01 | 0.11 | 1.82 |
| | N | 62 | 62 | 62 | 62 | 62 | 62 | 62 | 62 | 62 | 62 |
| Fall | min | 0.00 | 0.71 | 0.88 | 0.49 | 0.01 | 0.01 | 0.01 | 0.00 | 0.40 | 277.96 |
| | median | 0.02 | 2.04 | 1.67 | 1.25 | 0.03 | 0.02 | 0.02 | 0.00 | 0.62 | 290.19 |
| | max | 0.53 | 7.15 | 8.31 | 3.17 | 0.20 | 0.08 | 0.07 | 0.03 | 1.00 | 299.58 |
| | mean | 0.04 | 2.37 | 2.24 | 1.52 | 0.04 | 0.03 | 0.02 | 0.01 | 0.65 | 289.05 |
| | SD | 0.09 | 1.45 | 1.40 | 0.75 | 0.04 | 0.02 | 0.01 | 0.01 | 0.14 | 5.39 |
| | N | 40 | 40 | 40 | 40 | 40 | 40 | 40 | 40 | 40 | 40 |

[1] All the concentration values are expressed in $\mu g\ m^{-3}$.

[2] $TNH_3 = NH_3 + NH_4^+$; $THNO_3 = HNO_3 + NO_3^-$; $TH_2SO_4 = SO_4^{2-}$; $TNH_3$, $THNO_3$, $TH_2SO_4$ are all expressed as the equivalent concentration; T is temperature; RH is relative humidity; SD is standard deviation; N is the number of observations.

**Table S3**

The statistics of different precursor gases of $iPM_{2.5}$ by season at the YRK site in 2012-2016

| Season | | $Na^+$ | $TH_2SO_4$ | $TNH_3$ | $THNO_3$ | THCl | $Ca^{2+}$ | $K^+$ | $Mg^{2+}$ | RH | T (K) |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Winter | min | 0.00 | 0.33 | 0.74 | 0.61 | 0.01 | 0.00 | 0.01 | 0.00 | 0.31 | 265.13 |
| | median | 0.01 | 1.31 | 1.40 | 1.22 | 0.02 | 0.02 | 0.02 | 0.00 | 0.62 | 279.18 |
| | max | 0.35 | 3.78 | 4.23 | 2.50 | 0.04 | 0.03 | 0.04 | 0.01 | 0.85 | 287.25 |
| | mean | 0.04 | 1.56 | 1.53 | 1.41 | 0.02 | 0.01 | 0.02 | 0.00 | 0.61 | 278.22 |
| | SD | 0.07 | 0.97 | 0.69 | 0.55 | 0.01 | 0.01 | 0.01 | 0.00 | 0.15 | 4.83 |
| | N | 27 | 27 | 27 | 27 | 27 | 27 | 27 | 27 | 27 | 27 |
| Spring | min | 0.00 | 0.59 | 0.91 | 0.30 | 0.01 | 0.00 | 0.00 | 0.00 | 0.31 | 273.66 |
| | median | 0.02 | 1.58 | 2.06 | 1.13 | 0.02 | 0.02 | 0.02 | 0.01 | 0.66 | 291.94 |
| | max | 0.26 | 3.81 | 4.55 | 2.15 | 0.10 | 0.06 | 0.05 | 0.02 | 0.88 | 299.26 |
| | mean | 0.05 | 1.82 | 2.11 | 1.13 | 0.03 | 0.03 | 0.03 | 0.01 | 0.62 | 289.63 |
| | SD | 0.06 | 0.81 | 0.89 | 0.47 | 0.02 | 0.01 | 0.02 | 0.01 | 0.14 | 6.42 |
| | N | 30 | 30 | 30 | 30 | 30 | 30 | 30 | 30 | 30 | 30 |
| Summer | min | 0.01 | 0.70 | 1.10 | 0.39 | 0.01 | 0.00 | 0.00 | 0.00 | 0.44 | 290.01 |
| | median | 0.02 | 1.93 | 2.32 | 1.01 | 0.02 | 0.02 | 0.03 | 0.01 | 0.73 | 297.58 |
| | max | 0.20 | 4.15 | 10.69 | 2.15 | 0.07 | 0.09 | 0.07 | 0.03 | 0.87 | 303.83 |
| | mean | 0.03 | 2.05 | 2.66 | 1.11 | 0.03 | 0.03 | 0.03 | 0.01 | 0.71 | 297.56 |
| | SD | 0.04 | 0.92 | 1.62 | 0.49 | 0.01 | 0.02 | 0.02 | 0.01 | 0.11 | 2.27 |
| | N | 37 | 37 | 37 | 37 | 37 | 37 | 37 | 37 | 37 | 37 |
| Fall | min | 0.00 | 0.10 | 0.24 | 0.16 | 0.01 | 0.01 | 0.00 | 0.00 | 0.44 | 273.19 |
| | median | 0.02 | 1.29 | 1.91 | 0.97 | 0.02 | 0.02 | 0.02 | 0.00 | 0.66 | 290.73 |
| | max | 0.64 | 3.11 | 10.33 | 3.37 | 0.07 | 0.05 | 0.08 | 0.02 | 0.93 | 298.62 |
| | mean | 0.04 | 1.49 | 2.45 | 1.10 | 0.02 | 0.02 | 0.03 | 0.00 | 0.68 | 289.40 |
| | SD | 0.10 | 0.76 | 1.93 | 0.65 | 0.01 | 0.01 | 0.02 | 0.00 | 0.13 | 6.58 |
| | N | 39 | 39 | 39 | 39 | 39 | 39 | 39 | 39 | 39 | 39 |

[1] All the concentration values are expressed in $\mu g\ m^{-3}$.

[2] $TNH_3 = NH_3 + NH_4^+$; $THNO_3 = HNO_3 + NO_3^-$; $TH_2SO_4 = SO_4^{2-}$; $TNH_3$, $THNO_3$, $TH_2SO_4$ are all expressed as the equivalent concentration; T is temperature; RH is relative humidity; SD is standard deviation; N is the number of observations.

**Table S4**

The statistics of different precursor gases of $iPM_{2.5}$ by season at the JST site in 2010-2011

| Season | | $Na^+$ | $TH_2SO_4$ | $TNH_3$ | $THNO_3$ | THCl | $Ca^{2+}$ | $K^+$ | $Mg^{2+}$ | RH | T (K) |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Winter | min | 0.00 | 1.09 | 0.97 | 0.76 | 0.02 | 0.00 | 0.00 | 0.00 | 0.48 | 268.09 |
| | median | 0.03 | 1.86 | 1.82 | 1.29 | 0.03 | 0.03 | 0.02 | 0.00 | 0.61 | 276.09 |
| | max | 0.18 | 4.94 | 4.01 | 6.50 | 0.19 | 0.05 | 0.08 | 0.02 | 0.97 | 289.73 |
| | mean | 0.04 | 2.26 | 1.89 | 1.84 | 0.06 | 0.02 | 0.02 | 0.01 | 0.66 | 276.74 |
| | SD | 0.05 | 1.06 | 0.70 | 1.40 | 0.05 | 0.01 | 0.02 | 0.00 | 0.15 | 5.98 |
| | N | 17 | 17 | 17 | 17 | 17 | 17 | 17 | 17 | 17 | 17 |
| Spring | min | 0.01 | 1.00 | 1.50 | 0.87 | 0.01 | 0.01 | 0.01 | 0.00 | 0.39 | 275.99 |
| | median | 0.05 | 2.98 | 2.34 | 2.24 | 0.03 | 0.02 | 0.02 | 0.01 | 0.61 | 293.16 |
| | max | 0.14 | 7.65 | 3.89 | 3.72 | 0.15 | 0.03 | 0.11 | 0.02 | 0.80 | 300.87 |
| | mean | 0.06 | 3.30 | 2.44 | 2.14 | 0.04 | 0.02 | 0.04 | 0.01 | 0.61 | 289.56 |
| | SD | 0.05 | 1.62 | 0.69 | 0.84 | 0.03 | 0.01 | 0.03 | 0.00 | 0.09 | 7.17 |
| | N | 19 | 19 | 19 | 19 | 19 | 19 | 19 | 19 | 19 | 19 |
| Summer | min | 0.01 | 1.98 | 1.70 | 1.58 | 0.02 | 0.01 | 0.01 | 0.00 | 0.55 | 297.73 |
| | median | 0.02 | 3.77 | 2.60 | 2.25 | 0.03 | 0.03 | 0.03 | 0.01 | 0.64 | 300.86 |
| | max | 0.09 | 5.88 | 3.78 | 3.40 | 0.04 | 0.06 | 0.10 | 0.02 | 0.75 | 302.93 |
| | mean | 0.03 | 4.01 | 2.59 | 2.28 | 0.03 | 0.03 | 0.03 | 0.01 | 0.65 | 300.80 |
| | SD | 0.02 | 1.35 | 0.57 | 0.65 | 0.01 | 0.01 | 0.03 | 0.01 | 0.07 | 1.29 |
| | N | 17 | 17 | 17 | 17 | 17 | 17 | 17 | 17 | 17 | 17 |
| Fall | min | 0.01 | 0.88 | 1.18 | 0.66 | 0.02 | 0.01 | 0.01 | 0.00 | 0.47 | 283.32 |
| | median | 0.02 | 2.04 | 2.18 | 1.14 | 0.03 | 0.03 | 0.03 | 0.01 | 0.54 | 292.56 |
| | max | 0.05 | 4.48 | 3.92 | 2.77 | 0.05 | 0.05 | 0.11 | 0.01 | 0.70 | 300.74 |
| | mean | 0.02 | 2.15 | 2.31 | 1.42 | 0.03 | 0.03 | 0.04 | 0.01 | 0.55 | 292.20 |
| | SD | 0.01 | 1.09 | 0.78 | 0.70 | 0.01 | 0.01 | 0.03 | 0.00 | 0.07 | 5.83 |
| | N | 14 | 14 | 14 | 14 | 14 | 14 | 14 | 14 | 14 | 14 |

[1] All the concentration values are expressed in $\mu g\ m^{-3}$.

[2] $TNH_3 = NH_3 + NH_4^+$; $THNO_3 = HNO_3 + NO_3^-$; $TH_2SO_4 = SO_4^{2-}$; $TNH_3$, $THNO_3$, $TH_2SO_4$ are all expressed as the equivalent concentration; T is temperature; RH is relative humidity; SD is standard deviation; N is the number of observations.

**Table S5**

The statistics of different precursor gases of $iPM_{2.5}$ by season at the JST site in 2012-2016

| Season | | $Na^+$ | $TH_2SO_4$ | $TNH_3$ | $THNO_3$ | THCl | $Ca^{2+}$ | $K^+$ | $Mg^{2+}$ | RH | T (K) |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Winter | min | 0.01 | 0.57 | 0.95 | 0.58 | 0.02 | 0.01 | 0.01 | 0.00 | 0.37 | 271.33 |
| | median | 0.02 | 1.32 | 1.54 | 1.45 | 0.03 | 0.02 | 0.02 | 0.00 | 0.52 | 278.25 |
| | max | 0.13 | 3.38 | 2.85 | 3.92 | 0.39 | 0.05 | 0.07 | 0.02 | 0.95 | 290.89 |
| | mean | 0.04 | 1.65 | 1.69 | 1.73 | 0.07 | 0.02 | 0.03 | 0.01 | 0.58 | 279.74 |
| | SD | 0.03 | 0.85 | 0.58 | 0.96 | 0.09 | 0.01 | 0.02 | 0.00 | 0.18 | 5.37 |
| | N | 17 | 17 | 17 | 17 | 17 | 17 | 17 | 17 | 17 | 17 |
| Spring | min | 0.00 | 0.54 | 0.82 | 0.37 | 0.01 | 0.01 | 0.01 | 0.00 | 0.29 | 275.26 |
| | median | 0.03 | 1.67 | 1.96 | 1.08 | 0.02 | 0.02 | 0.03 | 0.01 | 0.57 | 290.19 |
| | max | 0.18 | 3.21 | 3.25 | 2.27 | 0.15 | 0.06 | 0.06 | 0.02 | 0.75 | 299.37 |
| | mean | 0.05 | 1.72 | 1.84 | 1.16 | 0.03 | 0.03 | 0.03 | 0.01 | 0.56 | 290.31 |
| | SD | 0.05 | 0.67 | 0.61 | 0.53 | 0.03 | 0.01 | 0.02 | 0.01 | 0.12 | 6.47 |
| | N | 29 | 29 | 29 | 29 | 29 | 29 | 29 | 29 | 29 | 29 |
| Summer | min | 0.01 | 0.69 | 0.86 | 0.73 | 0.01 | 0.01 | 0.01 | 0.00 | 0.42 | 296.19 |
| | median | 0.02 | 1.77 | 1.92 | 1.53 | 0.02 | 0.02 | 0.03 | 0.01 | 0.63 | 298.97 |
| | max | 0.11 | 3.99 | 2.87 | 3.71 | 0.05 | 0.07 | 0.23 | 0.02 | 0.85 | 305.40 |
| | mean | 0.03 | 2.02 | 1.96 | 1.70 | 0.02 | 0.03 | 0.05 | 0.01 | 0.63 | 299.53 |
| | SD | 0.02 | 0.94 | 0.58 | 0.67 | 0.01 | 0.01 | 0.05 | 0.01 | 0.10 | 2.27 |
| | N | 35 | 35 | 35 | 35 | 35 | 35 | 35 | 35 | 35 | 35 |
| Fall | min | 0.00 | 0.41 | 1.02 | 0.34 | 0.01 | 0.01 | 0.00 | 0.00 | 0.41 | 276.27 |
| | median | 0.02 | 1.30 | 1.86 | 1.00 | 0.02 | 0.03 | 0.03 | 0.01 | 0.63 | 293.00 |
| | max | 0.12 | 3.33 | 3.12 | 2.33 | 0.06 | 0.07 | 0.07 | 0.02 | 0.77 | 300.46 |
| | mean | 0.03 | 1.45 | 1.96 | 1.16 | 0.02 | 0.03 | 0.03 | 0.01 | 0.61 | 291.56 |
| | SD | 0.03 | 0.76 | 0.53 | 0.50 | 0.01 | 0.02 | 0.02 | 0.00 | 0.10 | 6.26 |
| | N | 36 | 36 | 36 | 36 | 36 | 36 | 36 | 36 | 36 | 36 |

[1] All the concentration values are expressed in $\mu g\ m^{-3}$.

[2] $TNH_3 = NH_3 + NH_4^+$; $THNO_3 = HNO_3 + NO_3^-$; $TH_2SO_4 = SO_4^{2-}$; $TNH_3$, $THNO_3$, $TH_2SO_4$ are all expressed as the equivalent concentration; T is temperature; RH is relative humidity; SD is standard deviation; N is the number of observations.

**Table S6**

The statistics of different precursor gases of $iPM_{2.5}$ by season at the CTR site in 2012-2016

| Season | | $Na^+$ | $TH_2SO_4$ | $TNH_3$ | $THNO_3$ | THCl | $Ca^{2+}$ | $K^+$ | $Mg^{2+}$ | RH | T (K) |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Winter | min | 0.01 | 0.36 | 0.21 | 0.19 | 0.01 | 0.01 | 0.00 | 0.00 | 0.37 | 271.98 |
| | median | 0.02 | 0.97 | 0.59 | 0.70 | 0.02 | 0.02 | 0.03 | 0.00 | 0.60 | 280.59 |
| | max | 0.42 | 4.40 | 1.48 | 2.22 | 0.09 | 0.06 | 0.08 | 0.02 | 0.94 | 290.03 |
| | mean | 0.05 | 1.41 | 0.65 | 0.99 | 0.02 | 0.02 | 0.03 | 0.01 | 0.61 | 279.95 |
| | SD | 0.08 | 1.02 | 0.32 | 0.61 | 0.02 | 0.01 | 0.02 | 0.00 | 0.15 | 4.47 |
| | N | 31 | 31 | 31 | 31 | 31 | 31 | 31 | 31 | 31 | 31 |
| Spring | min | 0.00 | 0.69 | 0.47 | 0.22 | 0.01 | 0.01 | 0.01 | 0.00 | 0.35 | 280.90 |
| | median | 0.02 | 1.69 | 0.84 | 0.66 | 0.02 | 0.02 | 0.04 | 0.01 | 0.58 | 291.83 |
| | max | 0.19 | 4.48 | 1.59 | 1.25 | 0.13 | 0.06 | 0.10 | 0.02 | 0.89 | 299.51 |
| | mean | 0.05 | 1.82 | 0.91 | 0.67 | 0.03 | 0.03 | 0.04 | 0.01 | 0.60 | 291.70 |
| | SD | 0.05 | 0.83 | 0.27 | 0.24 | 0.03 | 0.02 | 0.02 | 0.01 | 0.16 | 4.71 |
| | N | 34 | 34 | 34 | 34 | 34 | 34 | 34 | 34 | 34 | 34 |
| Summer | min | 0.00 | 0.69 | 0.24 | 0.29 | 0.01 | 0.00 | 0.01 | 0.00 | 0.51 | 296.19 |
| | median | 0.03 | 1.77 | 0.79 | 0.69 | 0.02 | 0.03 | 0.03 | 0.01 | 0.73 | 299.43 |
| | max | 0.19 | 3.95 | 1.48 | 1.60 | 0.58 | 0.21 | 0.08 | 0.05 | 0.87 | 303.32 |
| | mean | 0.04 | 1.93 | 0.82 | 0.70 | 0.04 | 0.04 | 0.04 | 0.01 | 0.72 | 299.62 |
| | SD | 0.04 | 0.77 | 0.31 | 0.28 | 0.08 | 0.04 | 0.02 | 0.01 | 0.09 | 1.56 |
| | N | 47 | 47 | 47 | 47 | 47 | 47 | 47 | 47 | 47 | 47 |
| Fall | min | 0.00 | 0.28 | 0.19 | 0.17 | 0.01 | 0.01 | 0.00 | 0.00 | 0.35 | 276.48 |
| | median | 0.02 | 1.26 | 0.71 | 0.72 | 0.02 | 0.03 | 0.03 | 0.01 | 0.67 | 292.91 |
| | max | 0.19 | 4.40 | 1.44 | 1.79 | 0.09 | 0.09 | 0.14 | 0.03 | 0.95 | 301.75 |
| | mean | 0.04 | 1.38 | 0.72 | 0.71 | 0.02 | 0.03 | 0.04 | 0.01 | 0.66 | 291.33 |
| | SD | 0.04 | 0.81 | 0.26 | 0.32 | 0.01 | 0.02 | 0.03 | 0.01 | 0.13 | 6.52 |
| | N | 62 | 62 | 62 | 62 | 62 | 62 | 62 | 62 | 62 | 62 |

[1] All the concentration values are expressed in $\mu g\ m^{-3}$.

[2] $TNH_3 = NH_3 + NH_4^+$; $THNO_3 = HNO_3 + NO_3^-$; $TH_2SO_4 = SO_4^{2-}$; $TNH_3$, $THNO_3$, $TH_2SO_4$ are all expressed as the equivalent concentration; T is temperature; RH is relative humidity; SD is standard deviation; N is the number of observations.

**Table S7**

The statistics of different precursor gases of $iPM_{2.5}$ by season at the BHM site in 2011

| Season | | $Na^+$ | $TH_2SO_4$ | $TNH_3$ | $THNO_3$ | THCl | $Ca^{2+}$ | $K^+$ | $Mg^{2+}$ | RH | T (K) |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Winter | min | 0.00 | 1.63 | 1.33 | 0.91 | 0.01 | 0.01 | 0.01 | 0.00 | 0.53 | 272.70 |
| | median | 0.04 | 2.47 | 3.21 | 2.31 | 0.33 | 0.05 | 0.05 | 0.01 | 0.61 | 279.40 |
| | max | 0.40 | 3.42 | 3.97 | 4.36 | 0.56 | 0.12 | 0.11 | 0.03 | 0.91 | 288.52 |
| | mean | 0.11 | 2.43 | 2.65 | 2.30 | 0.26 | 0.05 | 0.05 | 0.01 | 0.65 | 279.60 |
| | SD | 0.14 | 0.66 | 1.07 | 0.98 | 0.23 | 0.05 | 0.03 | 0.01 | 0.13 | 5.61 |
| | N | 9 | 9 | 9 | 9 | 9 | 9 | 9 | 9 | 9 | 9 |
| Spring | min | 0.01 | 1.35 | 1.61 | 0.94 | 0.02 | 0.02 | 0.02 | 0.00 | 0.45 | 288.03 |
| | median | 0.02 | 3.68 | 2.41 | 1.83 | 0.11 | 0.06 | 0.03 | 0.01 | 0.59 | 294.88 |
| | max | 0.22 | 5.38 | 5.20 | 2.60 | 0.50 | 0.08 | 0.15 | 0.03 | 0.72 | 301.94 |
| | mean | 0.08 | 3.55 | 2.77 | 1.80 | 0.15 | 0.05 | 0.06 | 0.02 | 0.58 | 295.16 |
| | SD | 0.09 | 1.64 | 1.16 | 0.65 | 0.15 | 0.02 | 0.05 | 0.01 | 0.08 | 4.78 |
| | N | 9 | 9 | 9 | 9 | 9 | 9 | 9 | 9 | 9 | 9 |
| Summer | min | 0.01 | 2.07 | 1.85 | 1.20 | 0.04 | 0.02 | 0.01 | 0.00 | 0.52 | 298.14 |
| | median | 0.03 | 4.05 | 3.23 | 1.95 | 0.08 | 0.04 | 0.05 | 0.01 | 0.70 | 301.47 |
| | max | 0.55 | 5.88 | 4.55 | 2.78 | 0.17 | 0.06 | 0.14 | 0.02 | 0.77 | 303.04 |
| | mean | 0.11 | 3.96 | 3.12 | 2.01 | 0.09 | 0.04 | 0.06 | 0.01 | 0.67 | 301.02 |
| | SD | 0.17 | 1.20 | 1.05 | 0.49 | 0.04 | 0.01 | 0.04 | 0.01 | 0.09 | 1.69 |
| | N | 9 | 9 | 9 | 9 | 9 | 9 | 9 | 9 | 9 | 9 |
| Fall | min | 0.00 | 1.65 | 1.43 | 0.72 | 0.03 | 0.02 | 0.01 | 0.00 | 0.46 | 285.59 |
| | median | 0.02 | 2.45 | 2.44 | 1.25 | 0.12 | 0.04 | 0.02 | 0.02 | 0.74 | 287.77 |
| | max | 0.13 | 4.49 | 2.89 | 1.93 | 0.16 | 0.16 | 0.13 | 0.03 | 0.87 | 291.71 |
| | mean | 0.05 | 2.77 | 2.29 | 1.31 | 0.11 | 0.06 | 0.04 | 0.02 | 0.70 | 288.42 |
| | SD | 0.05 | 1.08 | 0.61 | 0.59 | 0.05 | 0.06 | 0.05 | 0.01 | 0.16 | 3.00 |
| | N | 5 | 5 | 5 | 5 | 5 | 5 | 5 | 5 | 5 | 5 |

[1] All the concentration values are expressed in $\mu g\ m^{-3}$.

[2] $TNH_3 = NH_3 + NH_4^+$; $THNO_3 = HNO_3 + NO_3^-$; $TH_2SO_4 = SO_4^{2-}$; $TNH_3$, $THNO_3$, $TH_2SO_4$ are all expressed as the equivalent concentration; T is temperature; RH is relative humidity; SD is standard deviation; N is the number of observations.

**Table S8**

The statistics of different precursor gases of $iPM_{2.5}$ by season at the BHM site in 2012-2016

| Season | | $Na^+$ | $TH_2SO_4$ | $TNH_3$ | $THNO_3$ | THCl | $Ca^{2+}$ | $K^+$ | $Mg^{2+}$ | RH | T (K) |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Winter | min | 0.01 | 0.61 | 1.35 | 0.92 | 0.03 | 0.01 | 0.01 | 0.00 | 0.44 | 274.65 |
| | median | 0.02 | 1.26 | 1.61 | 1.27 | 0.10 | 0.02 | 0.02 | 0.01 | 0.62 | 281.76 |
| | max | 0.08 | 4.57 | 2.96 | 4.07 | 0.53 | 0.05 | 0.07 | 0.02 | 0.91 | 290.62 |
| | mean | 0.03 | 1.92 | 1.79 | 1.60 | 0.15 | 0.03 | 0.03 | 0.01 | 0.61 | 282.85 |
| | SD | 0.03 | 1.28 | 0.52 | 0.92 | 0.14 | 0.01 | 0.02 | 0.00 | 0.13 | 4.19 |
| | N | 11 | 11 | 11 | 11 | 11 | 11 | 11 | 11 | 11 | 11 |
| Spring | min | 0.01 | 1.05 | 1.20 | 0.55 | 0.02 | 0.01 | 0.01 | 0.00 | 0.44 | 282.14 |
| | median | 0.06 | 1.76 | 1.95 | 1.15 | 0.10 | 0.04 | 0.04 | 0.02 | 0.65 | 294.59 |
| | max | 0.18 | 3.95 | 3.19 | 2.13 | 0.52 | 0.18 | 0.07 | 0.03 | 0.88 | 298.40 |
| | mean | 0.07 | 2.00 | 2.01 | 1.22 | 0.14 | 0.05 | 0.04 | 0.02 | 0.63 | 292.62 |
| | SD | 0.05 | 0.89 | 0.61 | 0.46 | 0.13 | 0.05 | 0.02 | 0.01 | 0.13 | 5.09 |
| | N | 13 | 13 | 13 | 13 | 13 | 13 | 13 | 13 | 13 | 13 |
| Summer | min | 0.00 | 0.59 | 0.66 | 0.34 | 0.01 | 0.01 | 0.00 | 0.00 | 0.42 | 293.39 |
| | median | 0.03 | 2.26 | 2.17 | 1.20 | 0.04 | 0.03 | 0.04 | 0.01 | 0.63 | 299.67 |
| | max | 0.11 | 4.62 | 3.65 | 2.35 | 0.08 | 0.17 | 0.17 | 0.04 | 0.88 | 303.38 |
| | mean | 0.04 | 2.41 | 2.09 | 1.23 | 0.04 | 0.05 | 0.04 | 0.02 | 0.67 | 299.72 |
| | SD | 0.03 | 1.16 | 0.79 | 0.48 | 0.02 | 0.04 | 0.04 | 0.01 | 0.11 | 2.85 |
| | N | 25 | 25 | 25 | 25 | 25 | 25 | 25 | 25 | 25 | 25 |
| Fall | min | 0.01 | 0.40 | 0.83 | 0.44 | 0.02 | 0.03 | 0.02 | 0.00 | 0.42 | 280.01 |
| | median | 0.03 | 1.35 | 1.90 | 0.96 | 0.08 | 0.04 | 0.04 | 0.01 | 0.67 | 295.22 |
| | max | 0.15 | 4.63 | 4.27 | 2.10 | 0.64 | 0.15 | 0.10 | 0.07 | 0.73 | 301.70 |
| | mean | 0.05 | 1.98 | 2.17 | 1.02 | 0.13 | 0.05 | 0.05 | 0.02 | 0.63 | 293.08 |
| | SD | 0.04 | 1.27 | 0.97 | 0.39 | 0.16 | 0.03 | 0.02 | 0.01 | 0.10 | 6.58 |
| | N | 20 | 20 | 20 | 20 | 20 | 20 | 20 | 20 | 20 | 20 |

[1] All the concentration values are expressed in $\mu g\ m^{-3}$.

[2] $TNH_3 = NH_3 + NH_4^+$; $THNO_3 = HNO_3 + NO_3^-$; $TH_2SO_4 = SO_4^{2-}$; $TNH_3$, $THNO_3$, $TH_2SO_4$ are all expressed as the equivalent concentration; T is temperature; RH is relative humidity; SD is standard deviation; N is the number of observations.

**Table S9**

The statistics of different precursor gases of $iPM_{2.5}$ by season at the OAK site in 2010

| Season | | $Na^+$ | $TH_2SO_4$ | $TNH_3$ | $THNO_3$ | THCl | $Ca^{2+}$ | $K^+$ | $Mg^{2+}$ | RH | T (K) |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Winter | min | 0.01 | 0.56 | 0.42 | 0.27 | 0.02 | 0.01 | 0.00 | 0.00 | 0.33 | 270.37 |
| | median | 0.03 | 1.42 | 0.73 | 1.05 | 0.03 | 0.03 | 0.02 | 0.01 | 0.61 | 277.08 |
| | max | 0.13 | 4.84 | 2.05 | 1.91 | 0.08 | 0.03 | 0.10 | 0.02 | 0.85 | 290.14 |
| | mean | 0.04 | 1.85 | 0.87 | 1.05 | 0.04 | 0.02 | 0.04 | 0.01 | 0.63 | 279.03 |
| | SD | 0.04 | 1.28 | 0.44 | 0.44 | 0.02 | 0.00 | 0.03 | 0.01 | 0.16 | 5.83 |
| | N | 13 | 13 | 13 | 13 | 13 | 13 | 13 | 13 | 13 | 13 |
| Spring | min | 0.02 | 1.11 | 0.55 | 0.51 | 0.01 | 0.01 | 0.02 | 0.01 | 0.53 | 287.85 |
| | median | 0.09 | 2.96 | 1.30 | 0.71 | 0.06 | 0.02 | 0.04 | 0.01 | 0.72 | 293.77 |
| | max | 0.31 | 4.06 | 1.57 | 1.03 | 0.23 | 0.02 | 0.07 | 0.05 | 0.83 | 297.78 |
| | mean | 0.11 | 2.84 | 1.17 | 0.78 | 0.07 | 0.02 | 0.04 | 0.02 | 0.71 | 294.28 |
| | SD | 0.09 | 1.14 | 0.39 | 0.20 | 0.07 | 0.00 | 0.02 | 0.01 | 0.08 | 2.98 |
| | N | 9 | 9 | 9 | 9 | 9 | 9 | 9 | 9 | 9 | 9 |
| Summer | min | 0.02 | 2.14 | 0.56 | 0.43 | 0.02 | 0.01 | 0.01 | 0.00 | 0.66 | 300.21 |
| | median | 0.12 | 2.79 | 1.03 | 0.62 | 0.03 | 0.08 | 0.04 | 0.03 | 0.77 | 301.05 |
| | max | 0.21 | 5.26 | 1.59 | 0.85 | 0.10 | 0.13 | 0.05 | 0.05 | 0.85 | 303.66 |
| | mean | 0.11 | 3.09 | 1.06 | 0.63 | 0.04 | 0.07 | 0.03 | 0.03 | 0.77 | 301.56 |
| | SD | 0.08 | 1.10 | 0.44 | 0.15 | 0.03 | 0.05 | 0.01 | 0.02 | 0.07 | 1.24 |
| | N | 7 | 7 | 7 | 7 | 7 | 7 | 7 | 7 | 7 | 7 |
| Fall | min | 0.01 | 1.80 | 0.61 | 0.36 | 0.02 | 0.01 | 0.01 | 0.00 | 0.43 | 287.82 |
| | median | 0.04 | 2.32 | 1.28 | 0.65 | 0.02 | 0.02 | 0.03 | 0.01 | 0.73 | 293.35 |
| | max | 0.20 | 5.52 | 1.66 | 1.54 | 0.20 | 0.07 | 0.17 | 0.03 | 0.89 | 301.08 |
| | mean | 0.06 | 2.70 | 1.16 | 0.74 | 0.04 | 0.03 | 0.04 | 0.01 | 0.69 | 294.71 |
| | SD | 0.06 | 1.11 | 0.39 | 0.35 | 0.06 | 0.02 | 0.05 | 0.01 | 0.16 | 4.70 |
| | N | 10 | 10 | 10 | 10 | 10 | 10 | 10 | 10 | 10 | 10 |

[1] All the concentration values are expressed in $\mu g\ m^{-3}$.

[2] $TNH_3 = NH_3 + NH_4^+$; $THNO_3 = HNO_3 + NO_3^-$; $TH_2SO_4 = SO_4^{2-}$; $TNH_3$, $THNO_3$, $TH_2SO_4$ are all expressed as the equivalent concentration; T is temperature; RH is relative humidity; SD is standard deviation; N is the number of observations.

**Table S10**

The statistics of different precursor gases of $iPM_{2.5}$ by season at the OLF site in 2013-2016

| Season | | $Na^+$ | $TH_2SO_4$ | $TNH_3$ | $THNO_3$ | THCl | $Ca^{2+}$ | $K^+$ | $Mg^{2+}$ | RH | T (K) |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Winter | min | 0.00 | 0.22 | 0.43 | 0.30 | 0.01 | 0.01 | 0.01 | 0.00 | 0.38 | 268.30 |
| | median | 0.03 | 1.32 | 0.88 | 0.77 | 0.03 | 0.02 | 0.05 | 0.00 | 0.66 | 282.15 |
| | max | 0.41 | 4.16 | 2.52 | 1.96 | 0.48 | 0.04 | 0.15 | 0.05 | 0.96 | 295.25 |
| | mean | 0.07 | 1.50 | 1.02 | 0.85 | 0.06 | 0.02 | 0.06 | 0.01 | 0.68 | 282.58 |
| | SD | 0.10 | 0.95 | 0.57 | 0.47 | 0.10 | 0.01 | 0.04 | 0.01 | 0.15 | 5.77 |
| | N | 22 | 22 | 22 | 22 | 22 | 22 | 22 | 22 | 22 | 22 |
| Spring | min | 0.01 | 0.62 | 0.70 | 0.54 | 0.01 | 0.01 | 0.01 | 0.00 | 0.47 | 282.67 |
| | median | 0.06 | 1.52 | 1.18 | 0.93 | 0.04 | 0.03 | 0.04 | 0.01 | 0.72 | 294.65 |
| | max | 0.27 | 4.46 | 2.17 | 2.17 | 0.27 | 0.14 | 0.12 | 0.05 | 0.96 | 298.84 |
| | mean | 0.08 | 2.01 | 1.24 | 1.01 | 0.06 | 0.03 | 0.05 | 0.01 | 0.70 | 293.68 |
| | SD | 0.08 | 1.05 | 0.35 | 0.37 | 0.06 | 0.02 | 0.03 | 0.01 | 0.13 | 4.49 |
| | N | 23 | 23 | 23 | 23 | 23 | 23 | 23 | 23 | 23 | 23 |
| Summer | min | 0.02 | 0.86 | 0.43 | 0.38 | 0.02 | 0.01 | 0.01 | 0.00 | 0.69 | 297.99 |
| | median | 0.08 | 1.98 | 1.04 | 0.79 | 0.04 | 0.04 | 0.03 | 0.01 | 0.80 | 300.78 |
| | max | 0.27 | 4.20 | 1.78 | 2.31 | 0.16 | 0.12 | 0.11 | 0.05 | 0.87 | 302.99 |
| | mean | 0.09 | 2.14 | 1.00 | 0.88 | 0.06 | 0.05 | 0.04 | 0.02 | 0.79 | 300.94 |
| | SD | 0.06 | 0.97 | 0.36 | 0.40 | 0.04 | 0.03 | 0.02 | 0.01 | 0.05 | 1.18 |
| | N | 28 | 28 | 28 | 28 | 28 | 28 | 28 | 28 | 28 | 28 |
| Fall | min | 0.00 | 0.33 | 0.40 | 0.16 | 0.01 | 0.01 | 0.00 | 0.00 | 0.44 | 281.68 |
| | median | 0.05 | 1.30 | 0.79 | 0.51 | 0.02 | 0.02 | 0.04 | 0.01 | 0.73 | 295.37 |
| | max | 0.37 | 4.27 | 1.54 | 1.46 | 0.48 | 0.05 | 0.10 | 0.04 | 0.88 | 300.49 |
| | mean | 0.07 | 1.43 | 0.81 | 0.59 | 0.05 | 0.03 | 0.04 | 0.01 | 0.71 | 293.28 |
| | SD | 0.08 | 0.86 | 0.30 | 0.34 | 0.10 | 0.01 | 0.03 | 0.01 | 0.13 | 5.69 |
| | N | 23 | 23 | 23 | 23 | 23 | 23 | 23 | 23 | 23 | 23 |

[1] All the concentration values are expressed in $\mu g\ m^{-3}$.

[2] $TNH_3 = NH_3 + NH_4^+$; $THNO_3 = HNO_3 + NO_3^-$; $TH_2SO_4 = SO_4^{2-}$; $TNH_3$, $THNO_3$, $TH_2SO_4$ are all expressed as the equivalent concentration; T is temperature; RH is relative humidity; SD is standard deviation; N is the number of observations.

**Table S11**

The summary of aerosol pH at five sites in 2012 to 2016

| Site | Winter | Summer |
|---|---|---|
| YRK | 2.35 ± 0.53 | 1.73 ± 0.32 |
| JST | 3.49 ± 0.72 | 2.38 ± 0.18 |
| CTR | 2.40 ± 0.73 | 2.45 ± 0.15 |
| BHM | 3.17 ± 0.41 | 2.47 ± 0.17 |
| OLF | 2.96 ± 0.40 | 2.56 ± 0.12 |

**Table S12**

The summary of final MLR model coefficients at the JST site from 2010 to 2011

| Predictors | Coefficients | SE | t value | Pr > \|t\| |
|---|---|---|---|---|
| Intercept ($\beta_0$) | 0.08 | 0.04 | 1.97 | 0.054 |
| $SO_4^{2-}$ ($\beta_1$) | 0.31 | 0.01 | 32.79 | $< 2 \times 10^{-16}$ |
| $NO_3^-$ ($\beta_2$) | 0.25 | 0.03 | 7.91 | $5.57 \times 10^{-11}$ |
| $(NO_3^- - 0.66)^2$ ($\beta_3$) | 0.02 | 0.01 | 3.43 | 0.001 |
| $Mg^{2+}$ ($\beta_4$) | –6.69 | 2.95 | –2.27 | 0.026 |

[1]Residual standard error: 0.1104 on 62 degrees of freedom. Multiple R-squared: 0.96. Adjusted R-squared: 0.96. F-statistic: 397.4 on 4 and 62 D F, p-value: $< 2.2 \times 10^{-16}$.

**Table S13**

The summary of final MLR model coefficients at the JST site from 2012 to 2016

| Predictors | Coefficients | SE | t value | Pr > \|t\| |
|---|---|---|---|---|
| Intercept ($\beta_0$) | 0.1967 | 0.041 | 4.78 | $5.64 \times 10^{-6}$ |
| $SO_4^{2-}$ ($\beta_1$) | 0.35 | 0.0074 | 46.74 | $< 2 \times 10^{-16}$ |
| $NO_3^-$ ($\beta_2$) | 0.0052 | 0.0286 | 0.18 | 0.86 |
| $(NO_3^- - 0.45)^2$ ($\beta_3$) | 0.11 | 0.01 | 10.57 | $< 2 \times 10^{-16}$ |
| T ($\beta_4$) | –0.013 | 0.0021 | –6.08 | $1.88 \times 10^{-8}$ |
| $Mg^{2+}$ ($\beta_5$) | –1.15 | 1.68 | –0.69 | 0.49 |
| $(Mg^{2+} - 0.0077)^2$ ($\beta_6$) | –292.8 | 161.7 | –1.81 | 0.07 |
| $NH_3$ ($\beta_7$) | –0.06 | 0.027 | –2.22 | 0.03 |
| $(NH_3 - 1.31)^2$ ($\beta_8$) | –0.0297 | 0.014 | –2.16 | 0.03 |
| T:$NH_3$ ($\beta_9$) | 0.0055 | 0.0013 | 4.20 | $5.53 \times 10^{-5}$ |

[1]Residual standard error: 0.05489 on 107 degrees of freedom. Multiple R-squared: 0.97. Adjusted R-squared: 0.97. F-statistic: 462 on 9 and 107 DF, p-value: $< 2.2 \times 10^{-16}$.

**Table S14**

The summary of final MLR model coefficients at the BHM site in 2011

| Predictors | Coefficients | SE | t value | Pr > \|t\| |
|---|---|---|---|---|
| Intercept ($\beta_0$) | –0.0182 | 0.083 | –0.219 | 0.83 |
| $SO_4^{2-}$ ($\beta_1$) | 0.316 | 0.0132 | 23.95 | $5.86 \times 10^{-14}$ |
| $NO_3^-$ ($\beta_2$) | 0.489 | 0.0659 | 7.42 | $1.46 \times 10^{-6}$ |
| $(T - 18.23)^2$ ($\beta_3$) | 0.00052 | 0.000196 | 2.64 | 0.018 |
| $(Mg^{2+} - 0.014)^2$ ($\beta_4$) | –568.8 | 272.6 | –2.09 | 0.053 |
| $(NO_3^- - 0.68)^2$ ($\beta_5$) | –0.058 | 0.027 | –2.15 | 0.047 |
| $Ca^{2+}$ ($\beta_6$) | 3.85 | 0.904 | –4.26 | $5.96 \times 10^{-4}$ |
| $NH_3$ ($\beta_7$) | 0.067 | 0.0345 | 1.95 | 0.07 |
| $(NH_3 - 1.63)^2$ ($\beta_8$) | –0.063 | 0.0235 | –2.69 | 0.016 |
| $(Ca^{2+} - 0.052)^2$ ($\beta_9$) | 37.05 | 8.998 | 4.12 | 0.00081 |
| $(HNO_3 - 1.23)^2$ ($\beta_{10}$) | 0.110 | 0.041 | 2.60 | 0.019 |
| $(Cl^- - 0.16)^2$ ($\beta_{11}$) | 0.287 | 0.766 | 0.374 | 0.71 |
| T ($\beta_{12}$) | –0.00353 | 0.00484 | –0.729 | 0.48 |
| $Mg^{2+}$ ($\beta_{13}$) | –0.0106 | 2.55 | –0.004 | 0.997 |
| $HNO_3$ ($\beta_{14}$) | –0.0108 | 0.0458 | –0.235 | 0.82 |
| $Cl^-$ ($\beta_{15}$) | 0.18 | 0.249 | 0.721 | 0.48 |

[1]Residual standard error: 0.05312 on 16 degrees of freedom. Multiple R-squared: 0.99. Adjusted R-squared: 0.98. F-statistic: 126.5 on 15 and 16 DF, p-value: $1.008 \times 10^{-13}$.

**Table S15**

The summary of final MLR model coefficients at the BHM site from 2012 to 2016

| Predictors | Coefficients | SE | t value | Pr > \|t\| |
|---|---|---|---|---|
| Intercept ($\beta_0$) | 0.248 | 0.044 | 5.68 | $4.22 \times 10^{-7}$ |
| $SO_4^{2-}$ ($\beta_1$) | 0.292 | 0.024 | 12.09 | $< 2 \times 10^{-16}$ |
| T ($\beta_2$) | –0.012 | 0.002 | –6.17 | $6.28 \times 10^{-8}$ |
| $(NO_3^- - 0.34)^2$ ($\beta_3$) | 0.081 | 0.013 | 6.12 | $7.81 \times 10^{-8}$ |
| $(Cl^- - 0.102)^2$ ($\beta_4$) | 0.518 | 0.294 | 1.76 | 0.0833 |
| $Ca^{2+}$ ($\beta_5$) | –0.537 | 0.194 | –2.77 | 0.0075 |
| $NO_3^-$ ($\beta_6$) | 0.043 | 0.047 | 0.914 | 0.3642 |
| $Cl^-$ ($\beta_7$) | 0.037 | 0.124 | 0.301 | 0.7645 |
| $SO_4^{2-}$:T ($\beta_8$) | 0.0023 | 0.001 | 2.43 | 0.0181 |

[1]Residual standard error: 0.04904 on 60 degrees of freedom. Multiple R-squared: 0.99. Adjusted R-squared: 0.99. F-statistic: 641.6 on 8 and 60 D F, p-value: $< 2.2 \times 10^{-16}$.

**Table S16**

The summary of final MLR model coefficients at the CTR site from 2012 to 2016

| Predictors | Coefficients | SE | t value | Pr > \|t\| |
|---|---|---|---|---|
| Intercept ($\beta_0$) | 0.0049 | 0.012 | 0.403 | 0.687 |
| $SO_4^{2-}$ ($\beta_1$) | 0.32 | 0.0057 | 55.84 | $< 2 \times 10^{-16}$ |
| $NO_3^-$ ($\beta_2$) | 0.21 | 0.017 | 12.21 | $< 2 \times 10^{-16}$ |
| $Mg^{2+}$ ($\beta_3$) | –6.63 | 0.54 | –12.26 | $< 2 \times 10^{-16}$ |
| $NH_3$ ($\beta_4$) | 0.14 | 0.028 | 5.12 | $8.31 \times 10^{-7}$ |
| $(SO_4^{2-} - 1.59)^2$ ($\beta_5$) | –0.0169 | 0.0039 | –4.36 | $2.24 \times 10^{-5}$ |

[1]Residual standard error: 0.05129 on 168 degrees of freedom. Multiple R-squared: 0.96. Adjusted R-squared: 0.96. F-statistic: 921.2 on 5 and 168 DF, p-value: $< 2.2 \times 10^{-16}$.

**Table S17**

The summary of final MLR model coefficients at the OAK site in 2010

| Predictors | Coefficients | SE | t value | Pr > \|t\| |
|---|---|---|---|---|
| Intercept ($\beta_0$) | 0.103 | 0.054 | 1.89 | 0.068 |
| $SO_4^{2-}$ ($\beta_1$) | 0.36 | 0.014 | 26.28 | $< 2 \times 10^{-16}$ |
| T ($\beta_2$) | –0.0092 | 0.002 | –4.45 | 0.0001 |
| $Mg^{2+}$ ($\beta_3$) | –5.41 | 1.04 | –5.21 | $1.19 \times 10^{-5}$ |
| $(SO_4^{2-} - 2.47)^2$ ($\beta_4$) | –0.02 | 0.0076 | –2.62 | 0.013 |
| $NH_3$ ($\beta_5$) | 0.16 | 0.055 | 2.97 | 0.0056 |
| $NO_3^-$ ($\beta_6$) | 0.19 | 0.054 | 3.52 | 0.0013 |
| $(T - 17.46)^2$ ($\beta_7$) | –0.00028 | 0.00015 | –1.86 | 0.073 |

[1]Residual standard error: 0.06706 on 31 degrees of freedom. Multiple R-squared: 0.97. Adjusted R-squared: 0.97. F-statistic: 165.5 on 7 and 31 D F, p-value: $< 2.2 \times 10^{-16}$.

**Table S18**

The summary of final MLR model coefficients at the OLF site from 2013 to 2016

| Predictors | Coefficients | SE | t value | Pr > \|t\| |
|---|---|---|---|---|
| Intercept ($\beta_0$) | –0.0013 | 0.024 | –0.054 | 0.96 |
| $SO_4^{2-}$ ($\beta_1$) | 0.33 | 0.0075 | 43.69 | $< 2 \times 10^{-16}$ |
| $NO_3^-$ ($\beta_3$) | 0.35 | 0.061 | 5.74 | $1.36 \times 10^{-7}$ |
| $Mg^{2+}$ ($\beta_2$) | –4.69 | 0.88 | –5.30 | $8.78 \times 10^{-7}$ |
| $(Ca^{2+} - 0.03)^2$ ($\beta_4$) | –19.02 | 7.90 | –2.41 | 0.0181 |
| $(NO_3^- - 0.26)^2$ ($\beta_5$) | –0.283 | 0.0899 | –3.16 | 0.0022 |
| $(NH_3 - 0.45)^2$ ($\beta_6$) | 0.13 | 0.0819 | 1.59 | 0.1153 |
| $Ca^{2+}$ ($\beta_7$) | –0.522 | 0.602 | –0.868 | 0.3879 |
| $NH_3$ ($\beta_8$) | 0.0547 | 0.0425 | 1.288 | 0.2013 |

[1]Residual standard error: 0.0681 on 87 degrees of freedom. Multiple R-squared: 0.96. Adjusted R-squared: 0.96. F-statistic: 287.2 on 8 and 87 D F, p-value: $< 2.2 \times 10^{-16}$.

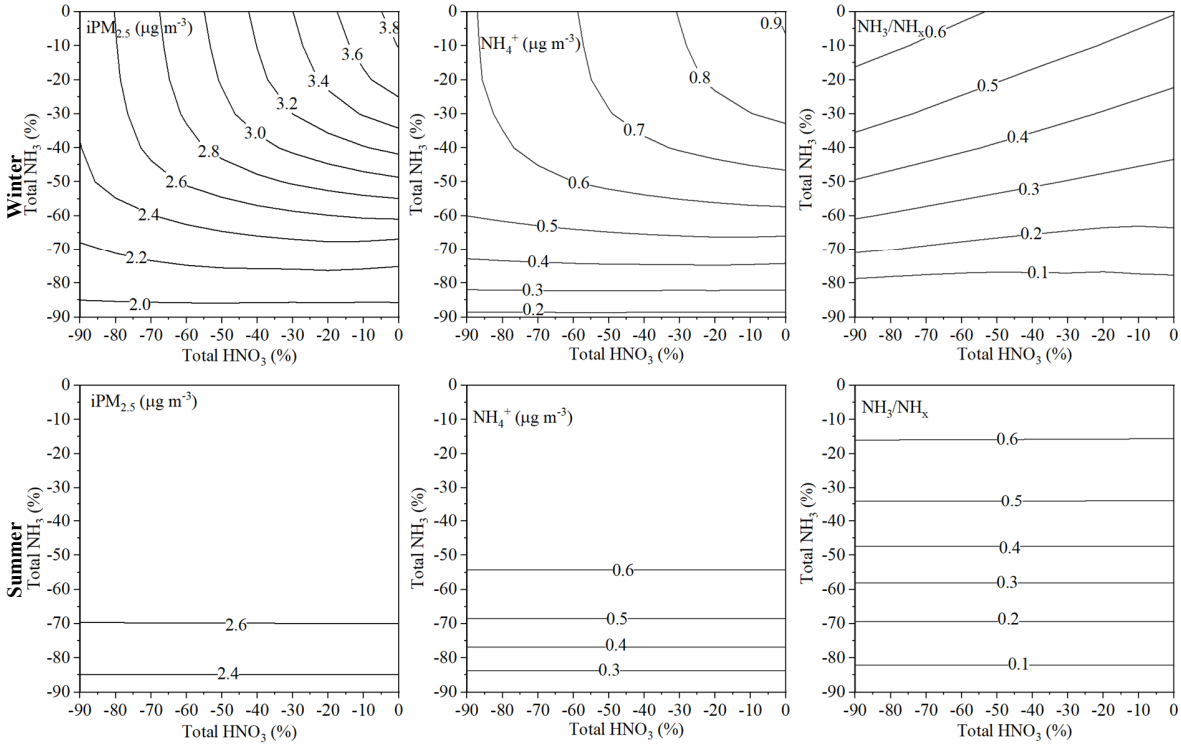


**Figure S1.** Responses of $iPM_{2.5}$, $NH_4^+$, and $NH_3/NH_x$ to the changes of $TNH_3$ and $THNO_3$ at the JST site in summer and winter of 2012-2016

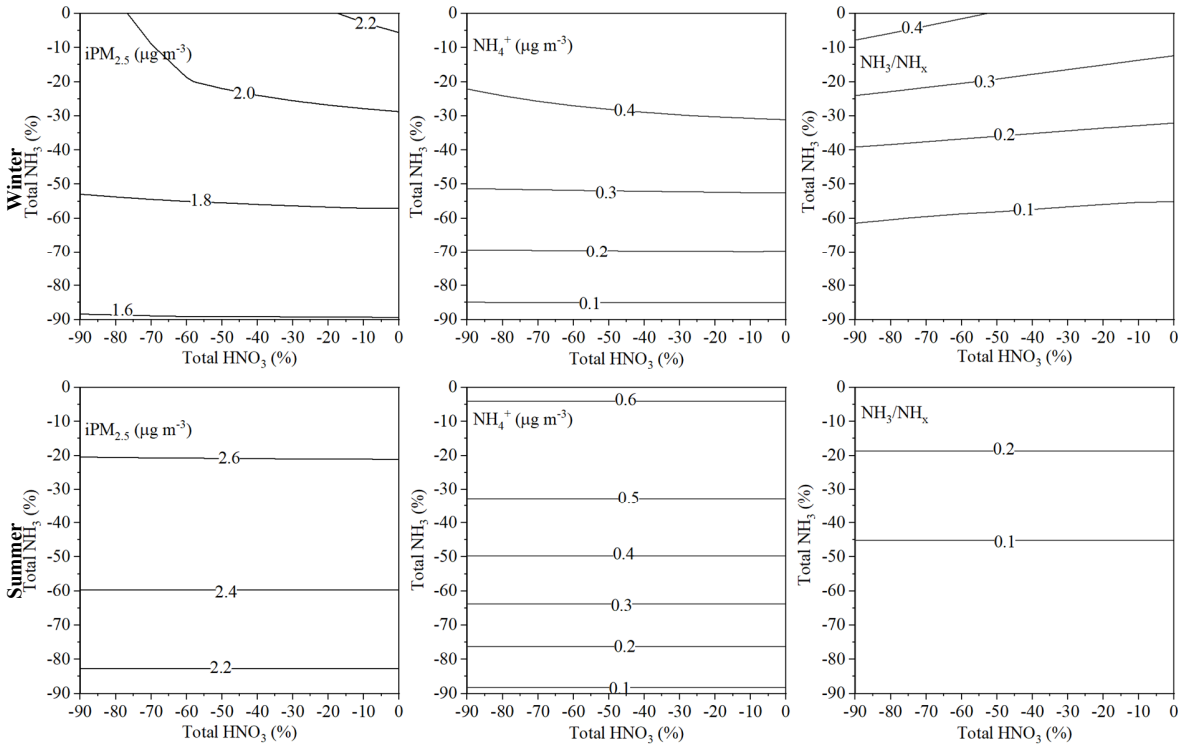


**Figure S2.** Responses of $iPM_{2.5}$, $NH_4^+$, and $NH_3/NH_x$ to the changes of $TNH_3$ and $THNO_3$ at the CTR site in summer and winter of 2012-2016

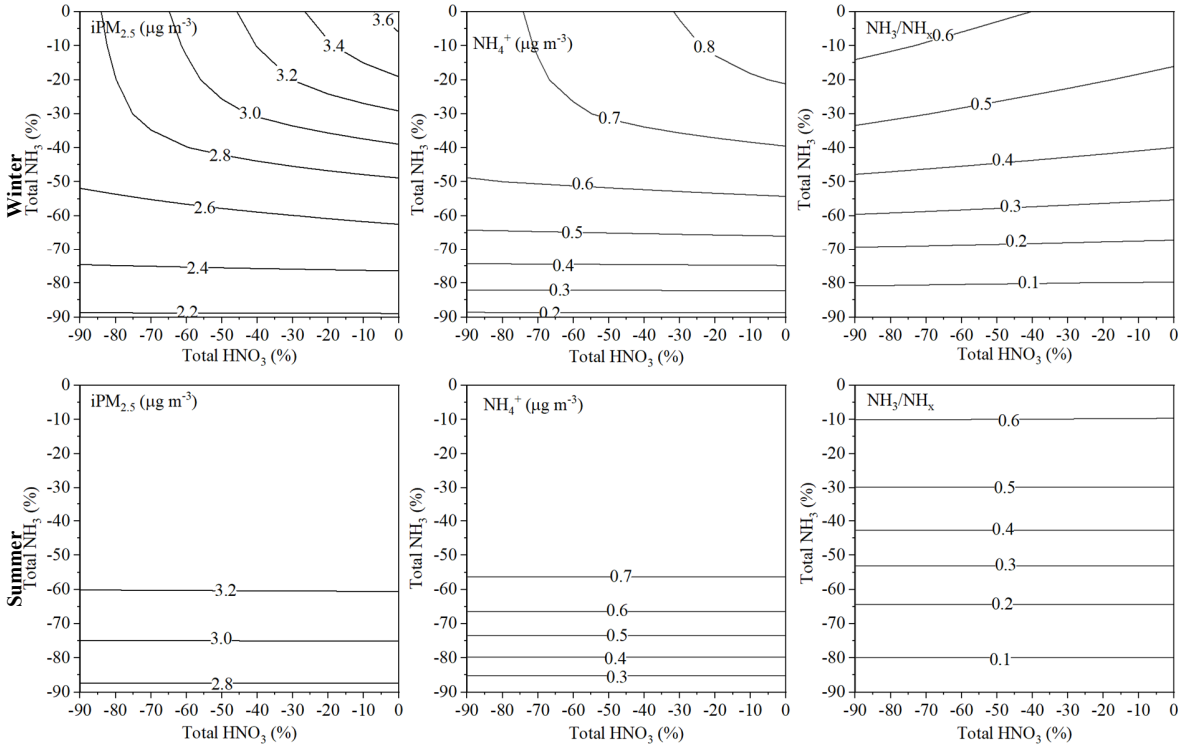


**Figure S3.** Responses of $iPM_{2.5}$, $NH_4^+$, and $NH_3/NH_x$ to the changes of $TNH_3$ and $THNO_3$ at the BHM site in summer and winter of 2012-2016

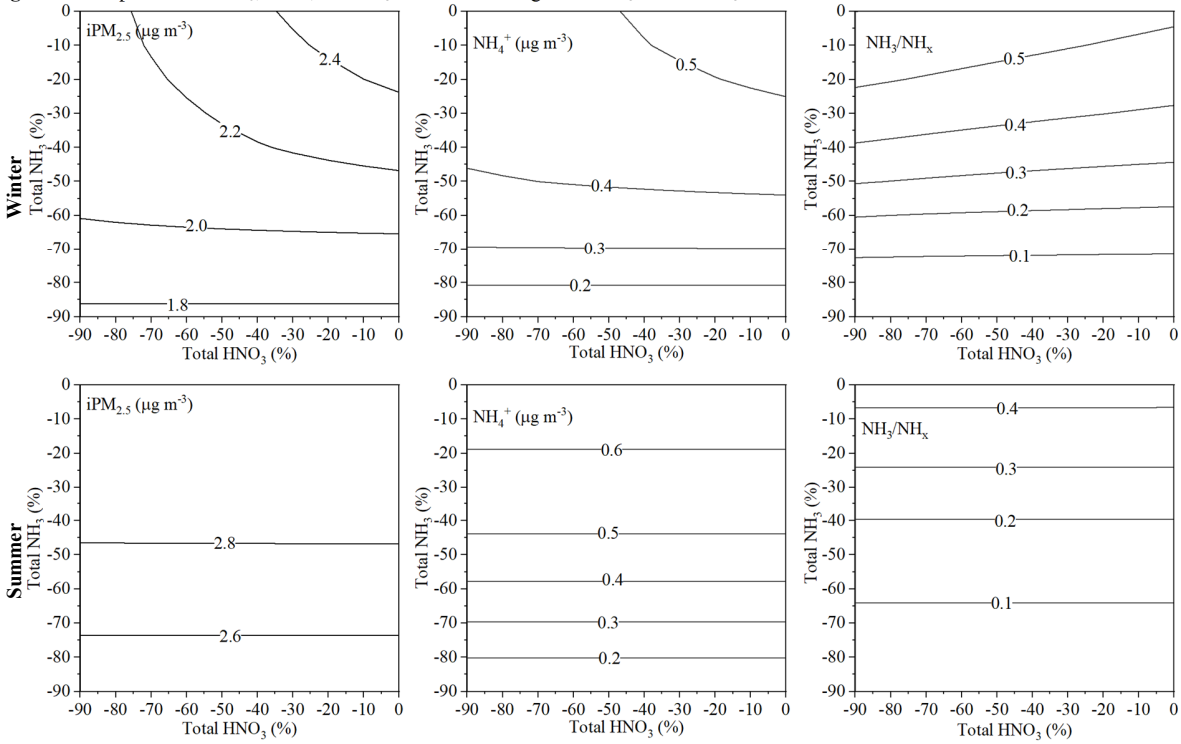


**Figure S4.** Responses of $iPM_{2.5}$, $NH_4^+$, and $NH_3/NH_x$ to the changes of $TNH_3$ and $THNO_3$ at the OLF site in summer and winter of 2013-2016

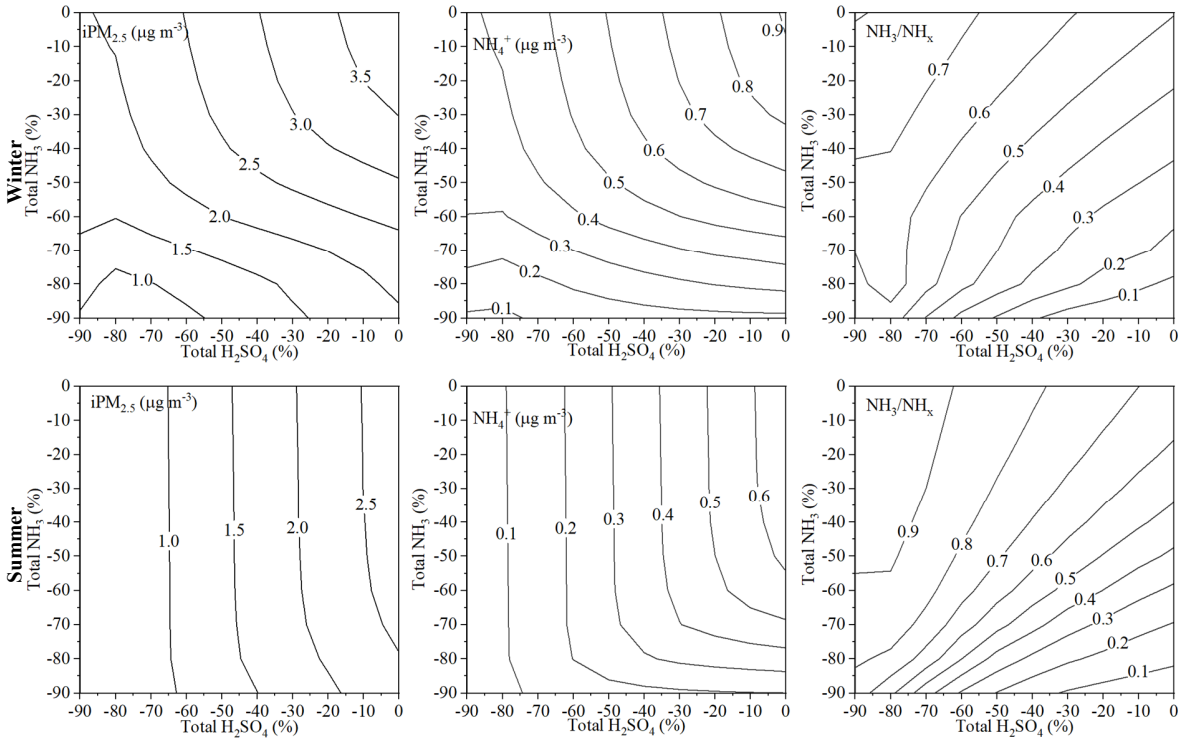


**Figure S5.** Responses of $iPM_{2.5}$, $NH_4^+$, and $NH_3/NH_x$ to the changes of $TNH_3$ and $TH_2SO_4$ at the JST site in summer and winter of 2012-2016

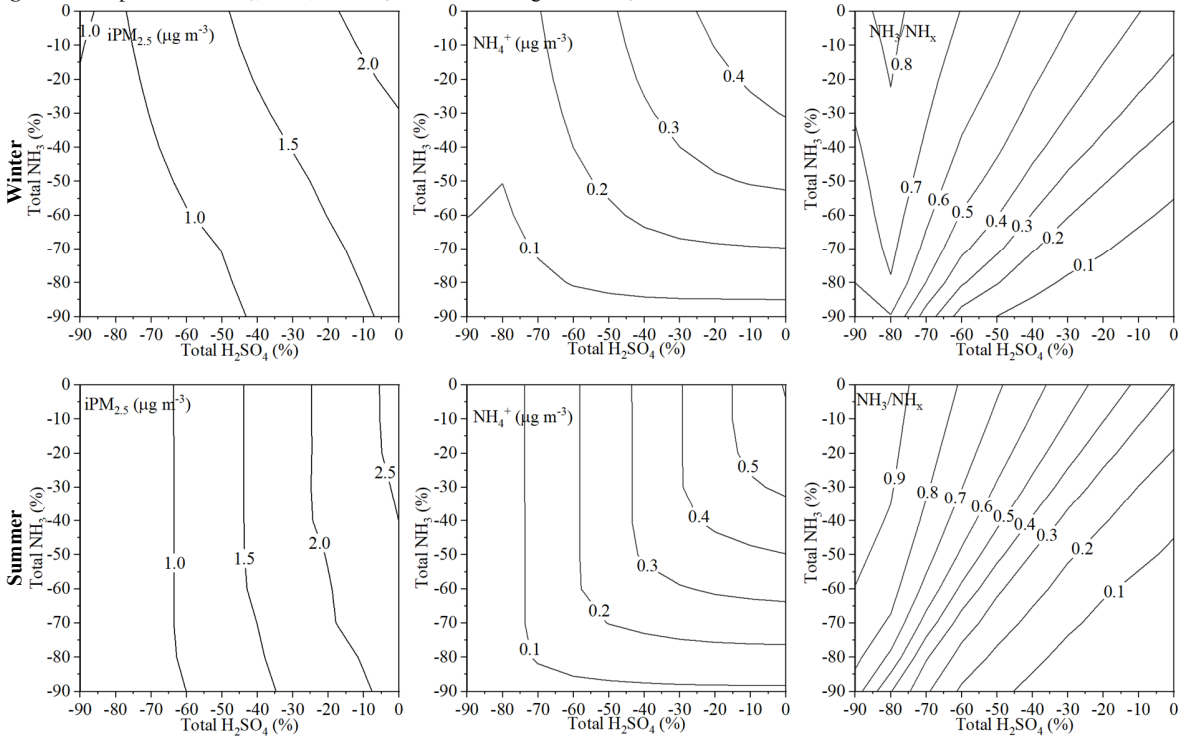


**Figure S6.** Responses of $iPM_{2.5}$, $NH_4^+$, and $NH_3/NH_x$ to the changes of $TNH_3$ and $TH_2SO_4$ at the CTR site in summer and winter of 2012-2016

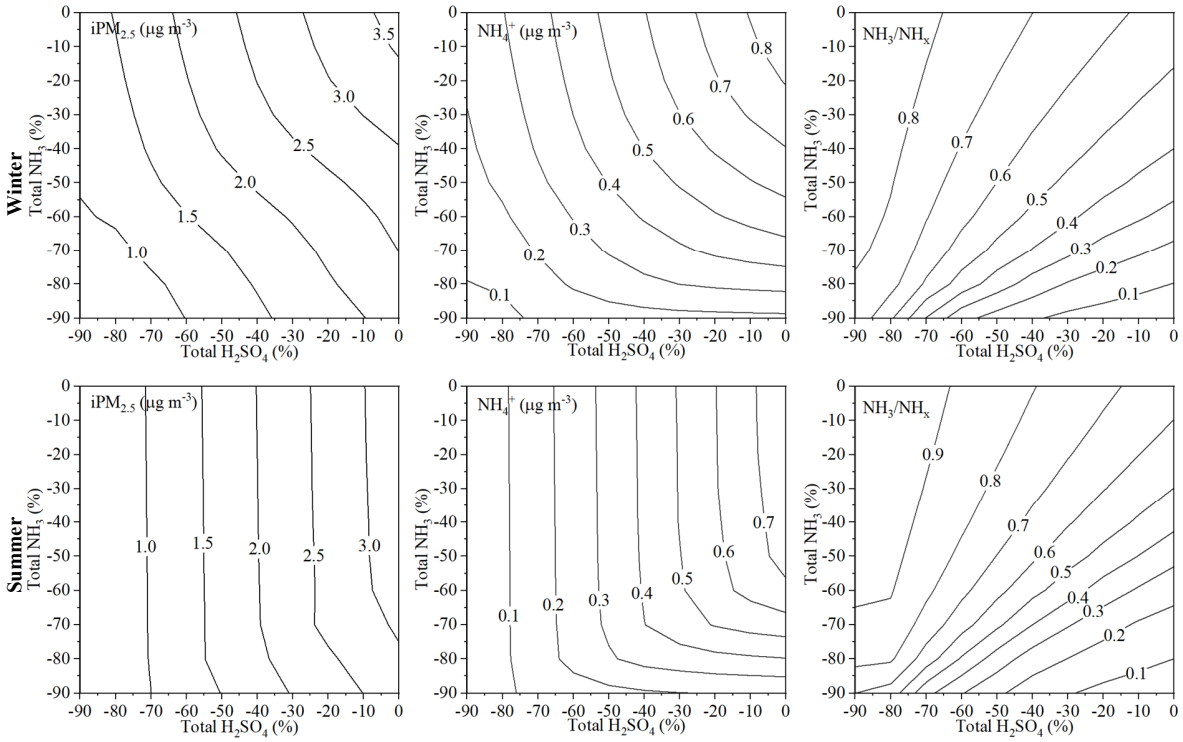


**Figure S7.** Responses of $iPM_{2.5}$, $NH_4^+$, and $NH_3/NH_x$ to the changes of $TNH_3$ and $TH_2SO_4$ at the BHM site in summer and winter of 2012-2016

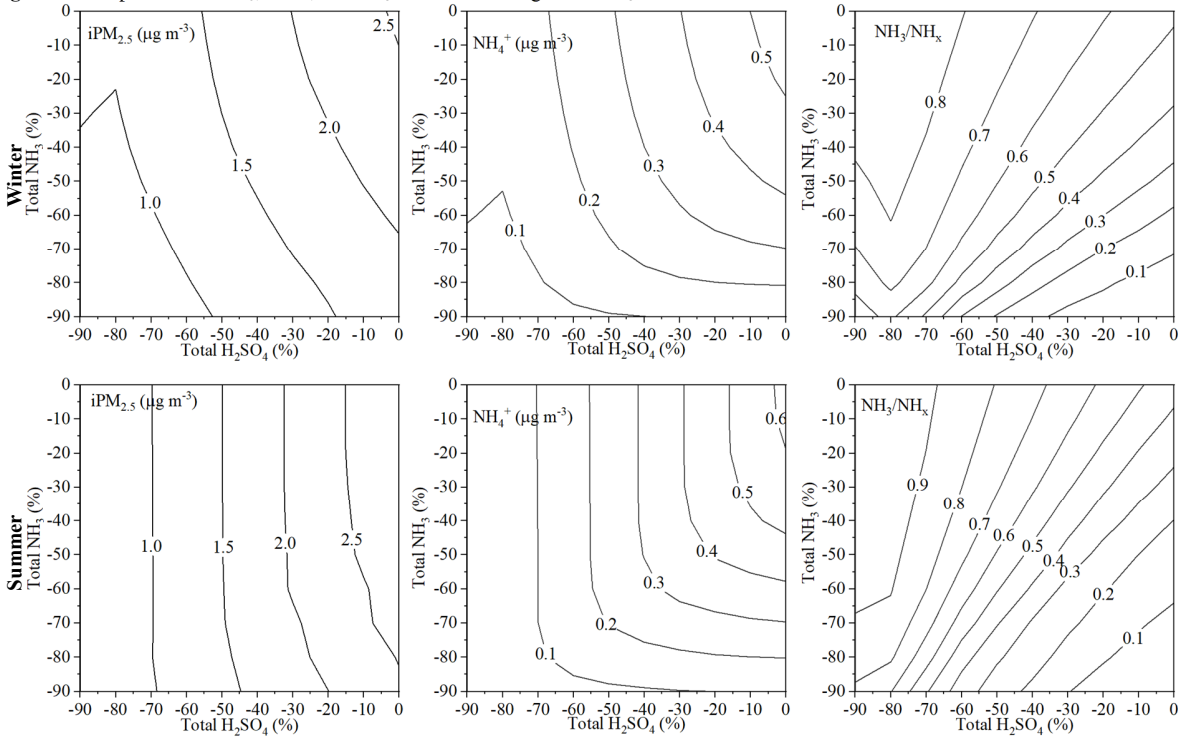


**Figure S8.** Responses of $iPM_{2.5}$, $NH_4^+$, and $NH_3/NH_x$ to the changes of $TNH_3$ and $TH_2SO_4$ at the OLF site in summer and winter of 2013-2016

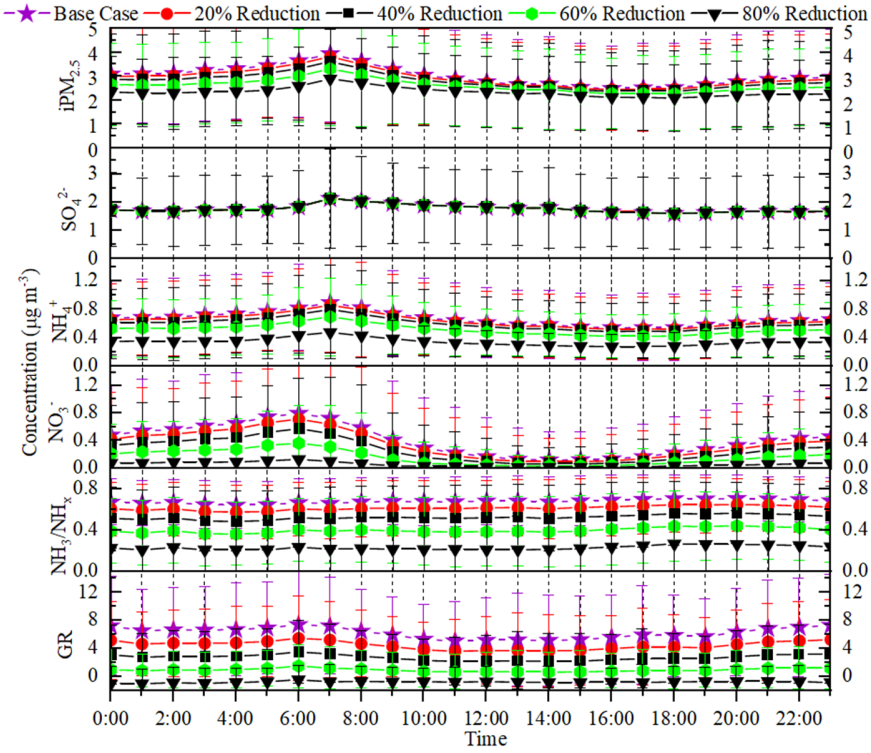


**Figure S9.** Responses of $iPM_{2.5}$, $SO_4^{2-}$, $NH_4^+$, $NO_3^-$, $NH_3/NH_x$, and GR to the reductions in $TNH_3$ at the BHM site in 2012-2016

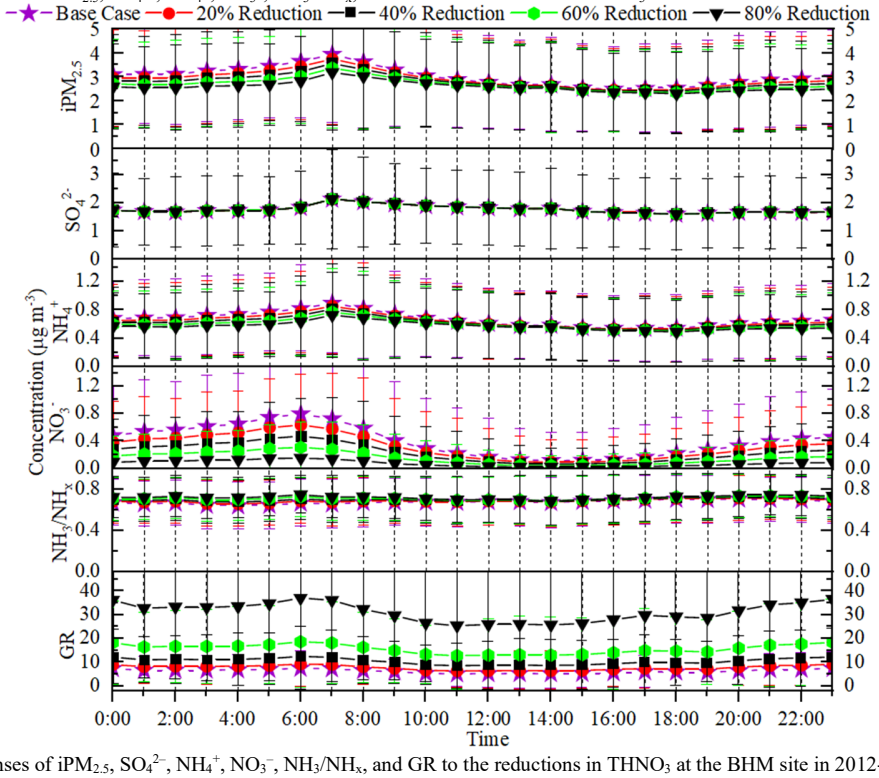


**Figure S10.** Responses of $iPM_{2.5}$, $SO_4^{2-}$, $NH_4^+$, $NO_3^-$, $NH_3/NH_x$, and GR to the reductions in $THNO_3$ at the BHM site in 2012-2016

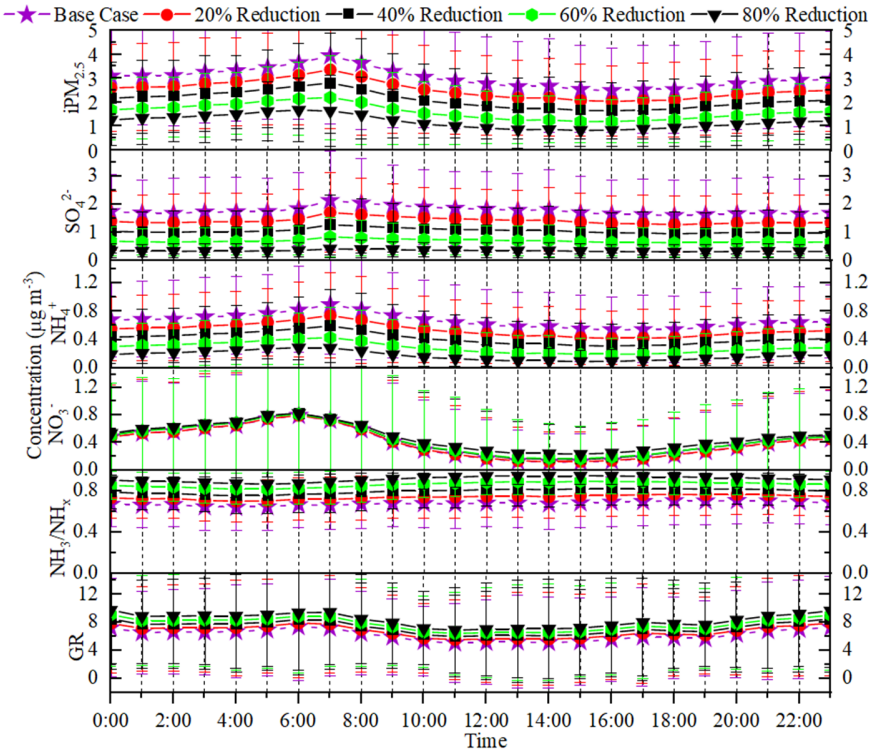


**Figure S11.** Responses of $iPM_{2.5}$, $SO_4^{2-}$, $NH_4^+$, $NO_3^-$, $NH_3/NH_x$, and GR to the reductions in $TH_2SO_4$ at the BHM site in 2012-2016

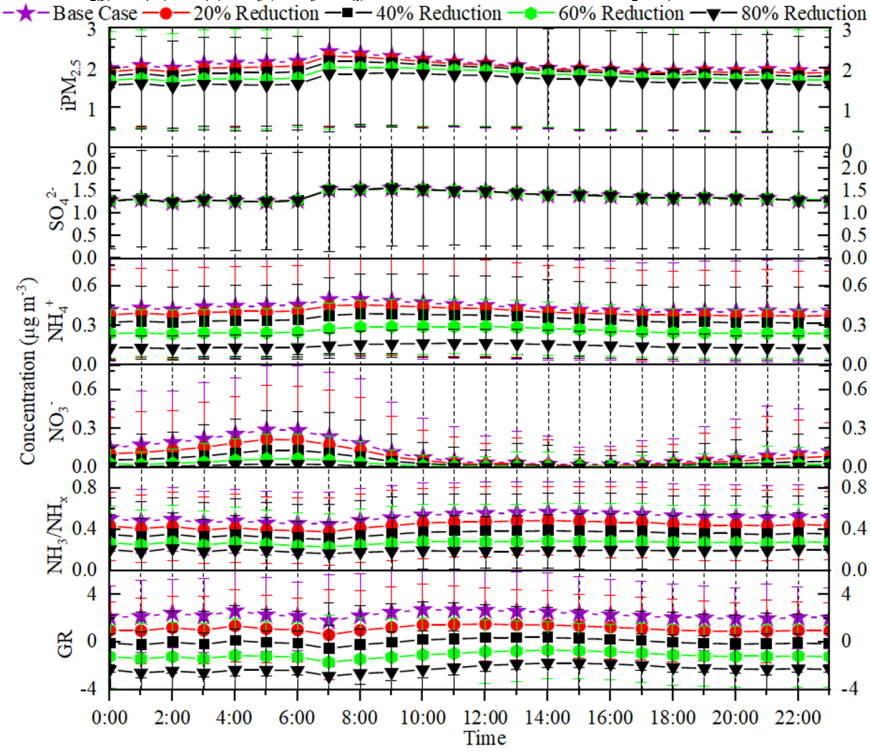


**Figure S12.** Responses of $iPM_{2.5}$, $SO_4^{2-}$, $NH_4^+$, $NO_3^-$, $NH_3/NH_x$, and GR to the reductions in $TNH_3$ at the CTR site in 2012-2016

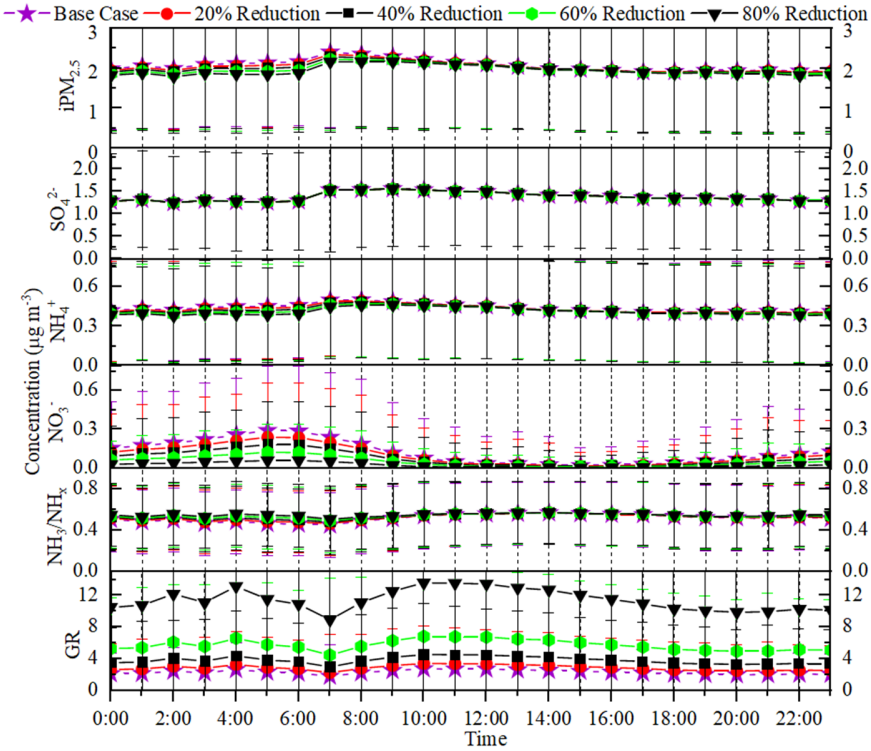


**Figure S13.** Responses of $iPM_{2.5}$, $SO_4^{2-}$, $NH_4^+$, $NO_3^-$, $NH_3/NH_x$, and GR to the reductions in $THNO_3$ at the CTR site in 2012-2016

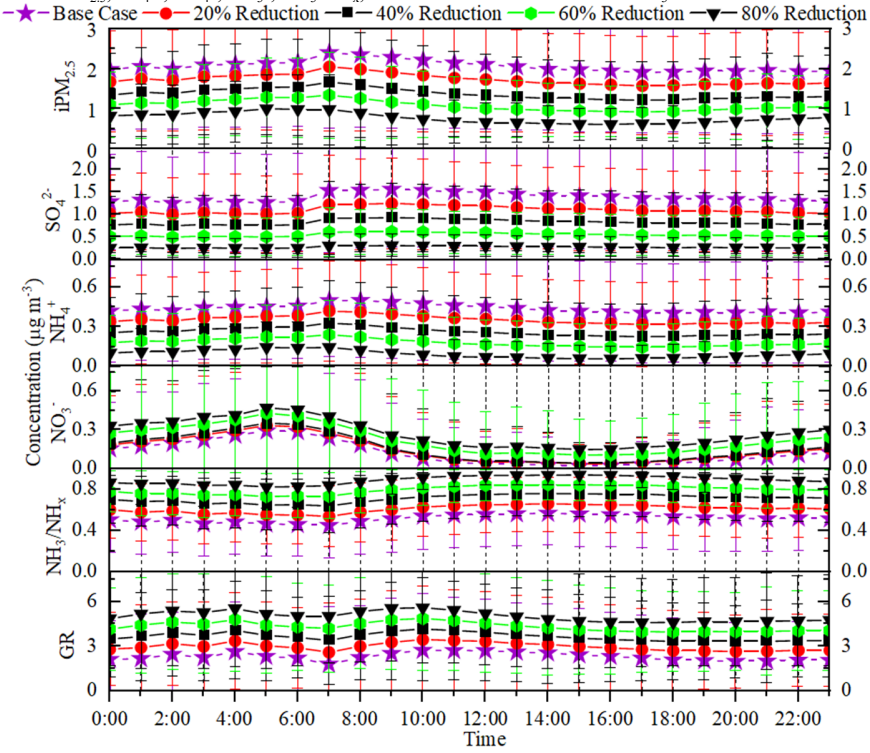


**Figure S14.** Responses of $iPM_{2.5}$, $SO_4^{2-}$, $NH_4^+$, $NO_3^-$, $NH_3/NH_x$, and GR to the reductions in $TH_2SO_4$ at the CTR site in 2012-2016

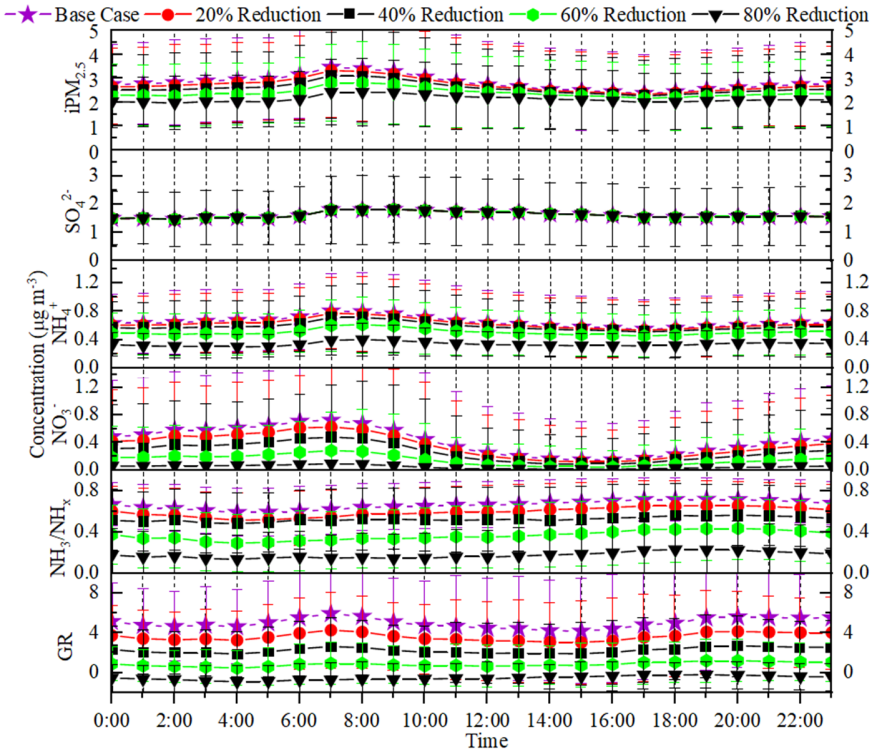


**Figure S15.** Responses of $iPM_{2.5}$, $SO_4^{2-}$, $NH_4^+$, $NO_3^-$, $NH_3/NH_x$, and GR to the reductions in $TNH_3$ at the JST site in 2012-2016

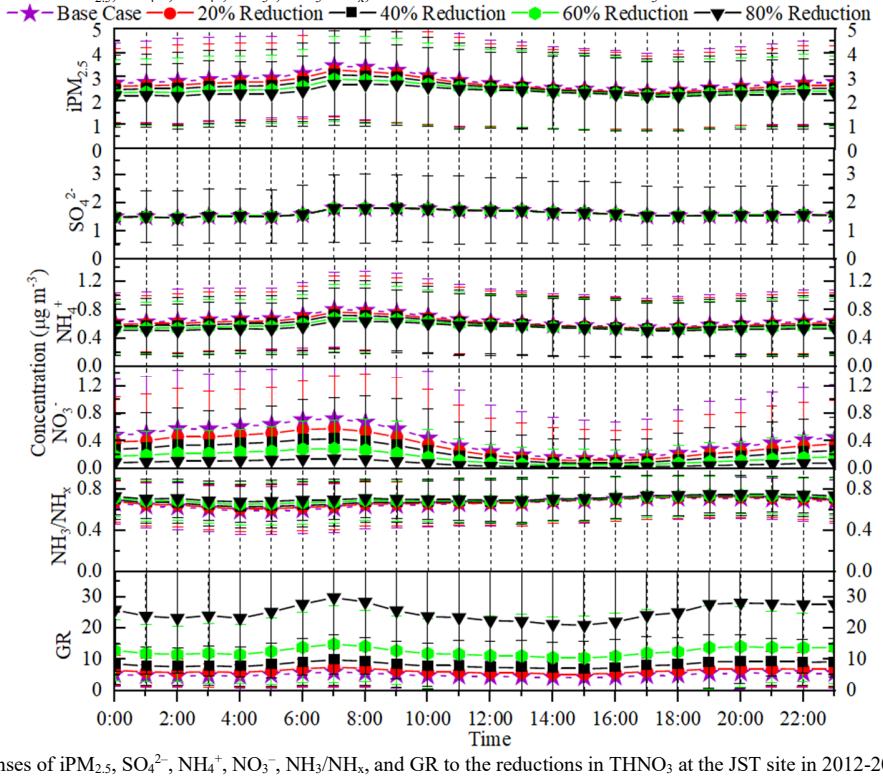


**Figure S16.** Responses of $iPM_{2.5}$, $SO_4^{2-}$, $NH_4^+$, $NO_3^-$, $NH_3/NH_x$, and GR to the reductions in $THNO_3$ at the JST site in 2012-2016

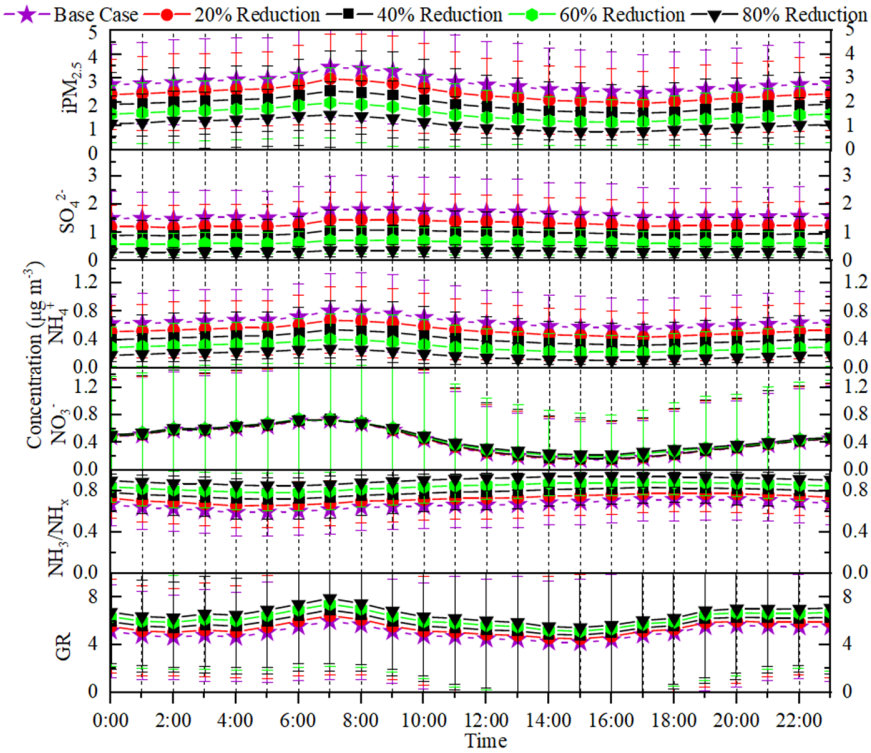


**Figure S17.** Responses of $iPM_{2.5}$, $SO_4^{2-}$, $NH_4^+$, $NO_3^-$, $NH_3/NH_x$, and GR to the reductions in $TH_2SO_4$ at the JST site in 2012-2016

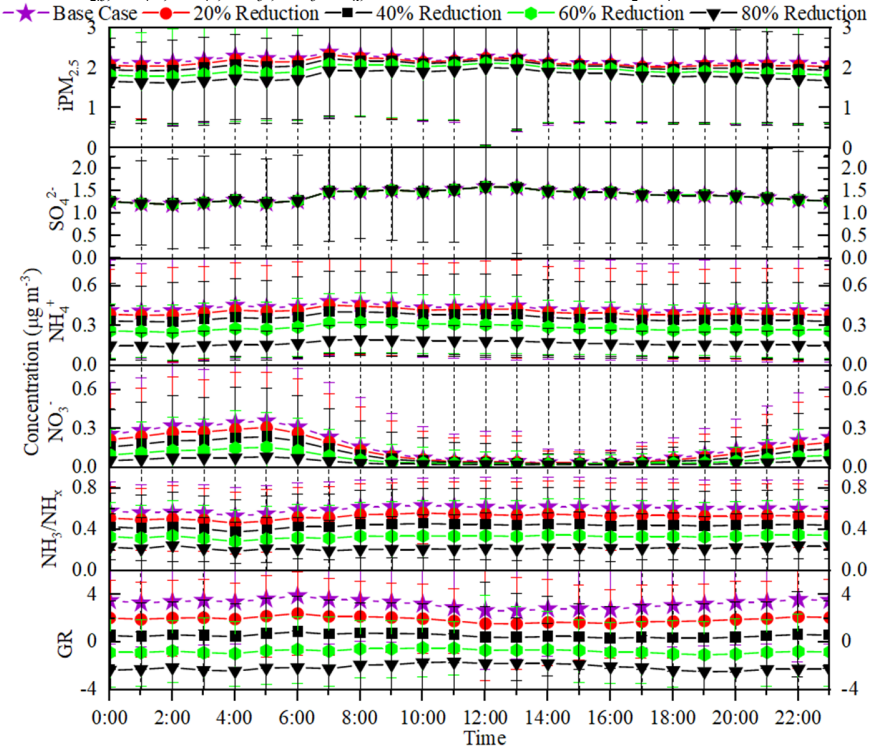


**Figure S18.** Responses of $iPM_{2.5}$, $SO_4^{2-}$, $NH_4^+$, $NO_3^-$, $NH_3/NH_x$, and GR to the reductions in $TNH_3$ at the OLF site in 2013-2016

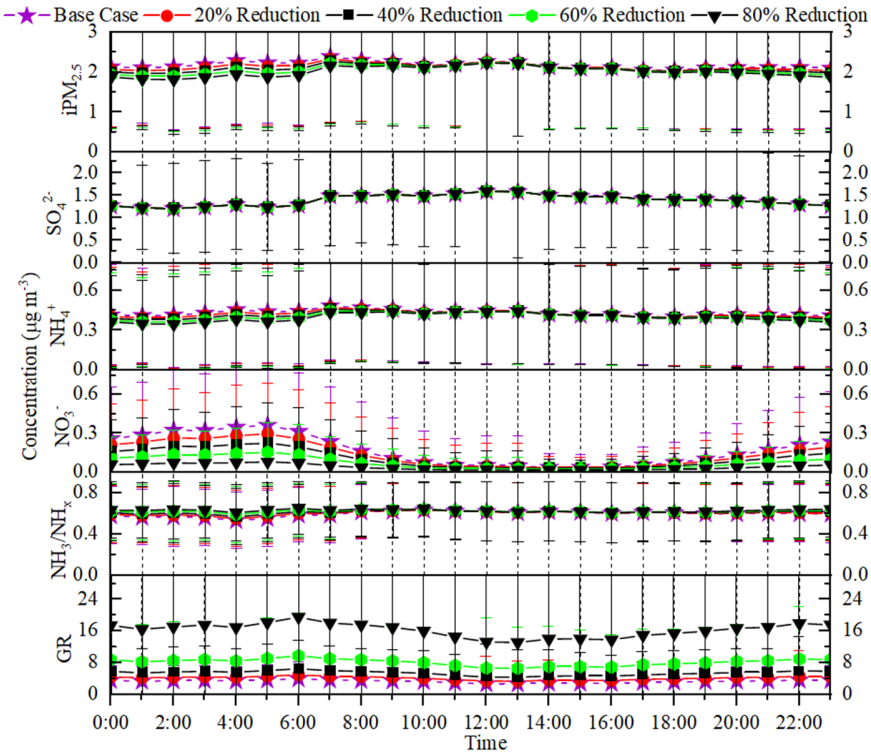


**Figure S19.** Responses of $iPM_{2.5}$, $SO_4^{2-}$, $NH_4^+$, $NO_3^-$, $NH_3/NH_x$, and GR to the reductions in $THNO_3$ at the OLF site in 2013-2016

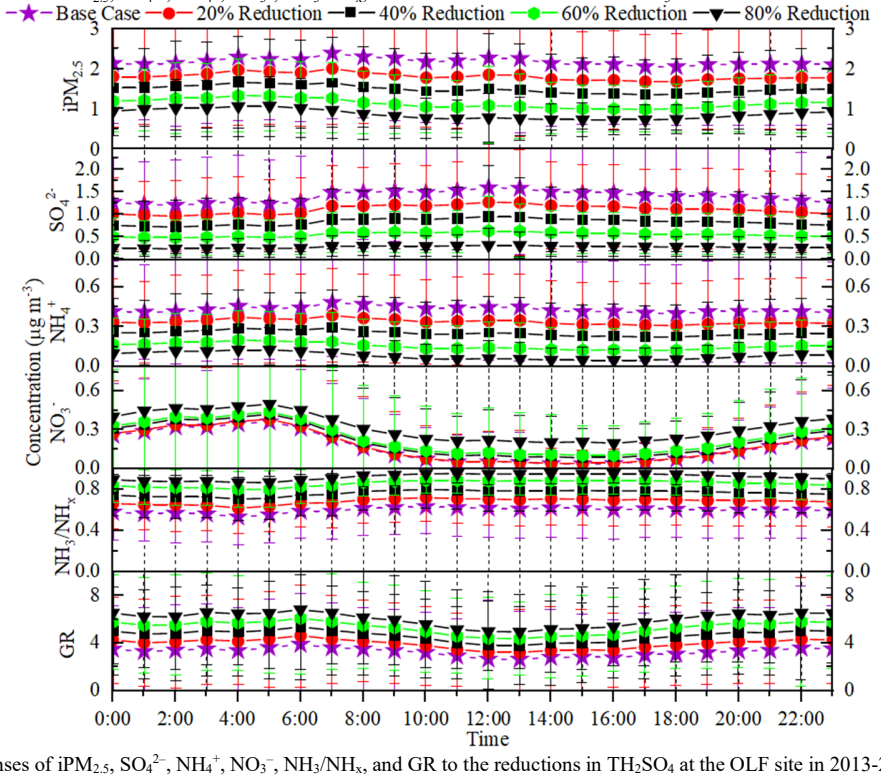


**Figure S20.** Responses of $iPM_{2.5}$, $SO_4^{2-}$, $NH_4^+$, $NO_3^-$, $NH_3/NH_x$, and GR to the reductions in $TH_2SO_4$ at the OLF site in 2013-2016