Supplementary Information for:

**Composition and oxidation state of sulfur in atmospheric particulate matter**

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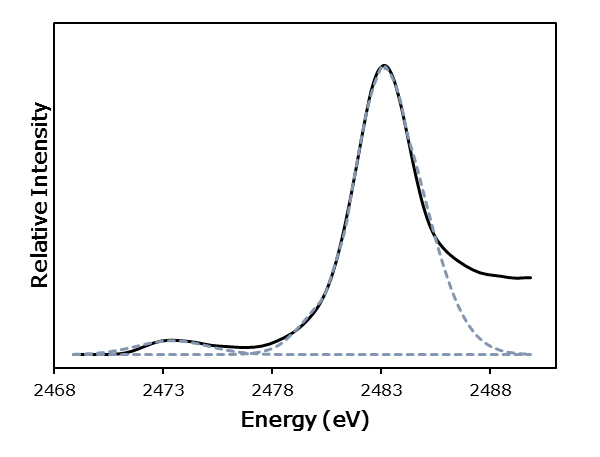
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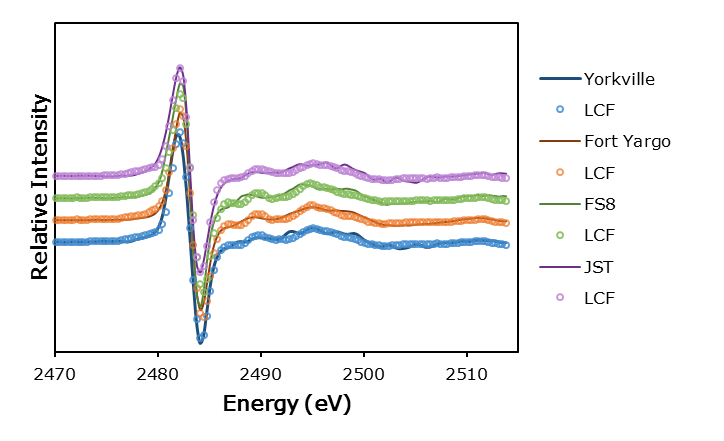
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**Table S1** Summary of ambient PM2.5 collection

|  |  |  |  |
| --- | --- | --- | --- |
| **Location** | **Description** | **Date** | **No. of Individual Particles Examined** |
| **Fire Station 8** | Urban | 8.6.09 | - |
| 8.26.09 | - |
| 6.14.11 | - |
| 6.17.11 | - |
| 8.2.11 | - |
| 8.11.11 | 5 |
| 8.24.11 | - |
| **South DeKalb** | Urban | 8.5.09 | - |
| 8.19.11 | - |
| 8.24.11 | 4 |
| 2.4.13 | - |
| **Jefferson Street** | Urban Background | 5.24.12 | 2 |
| 8.6.12 | - |
| **Fort Yargo** | Rural | 9.17.08 | 5 |
| 5.12.09 | - |
| 6.10.11 | 5 |
| 6.13.11 | - |
| 8.18.11 | - |
| **Yorkville** | Rural Background | 6.20.12 | 2 |
| 6.22.12 | - |
| 6.29.12 | - |

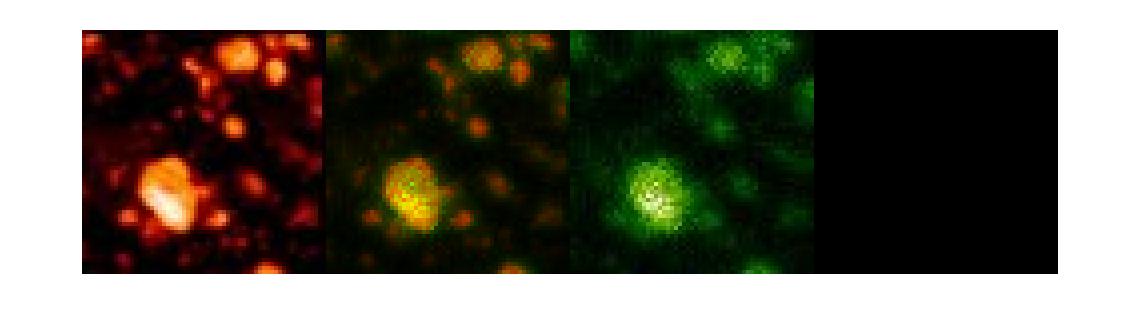


**Figure S1:** Gaussian peak fitting was used to determine the relative abundance of the S0 and S+VI oxidation state in the aerosol samples. Athena’s peak fitting protocol was used to fit Gaussian curves (dotted line) to the S-NEXFS spectrum (solid line) and to determine the area under the Gaussian curves.



**Figure S2:** Linear combination fits of representative bulk samples. The first derivative was used to fit ambient aerosol samples (solid lines) with a database of sulfate standards. The resulting linear combination fits are shown with open circles.

**Figure S3:** X-ray fluorescence map of sulfur for two ambient particulate matter samples. The sulfur elemental density maps are scaled from black, lowest intensity, to white for the highest intensity. The maximum intensity value (in raw detector counts) for the elemental map is shown in the upper corner, with corresponding scale bars on the side of the elemental density maps. For both samples, S+VI (red) is concentrated in the center of the particle, where the particle mass is highest. S0 (green) has a more varied spatial distribution, but is generally concentrated in one specific area. An overlay of the S0 and S+VI elemental density is shown in the center.

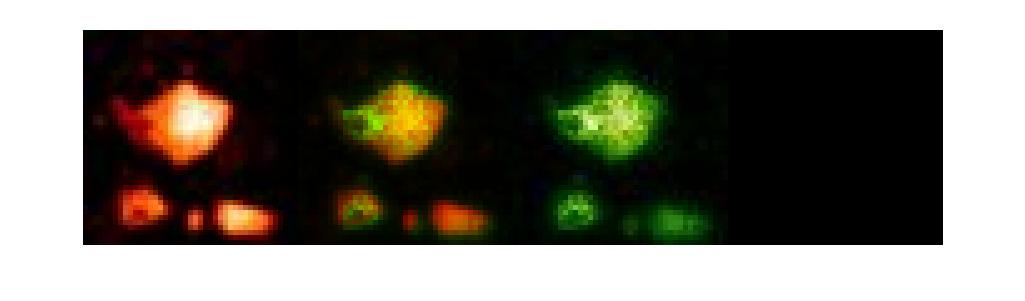


0.0278

0.00219

**S+VI**

**S0**



0.0373

0.00420

**S+VI**

**S0**

**Table S2 Composition of Sulfate from Bulk Ambient Aerosols and Emission Sources**

|  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Sampling Location** | **Ammonium Sulfate** | | **Cu(II) Sulfate** | | **Gypsum** | | **Iron(III) Sulfate** | | **Potassium Sulfate** | | ***R* factor** |
| % | ± | % | ± | % | ± | % | ± | % | ± |
| **Yorkville** | 61.3 | 8.5 | - | - | - | - | 38.7 | 8.5 | - | - | 0.096963 |
| **Fort Yargo** | 83.9 | 7.4 | - | - | - | - | 16.1 | 7.4 | - | - | 0.0767302 |
| **Fire Station 8** | 86.5 | 6.5 | 13.5 | 16.3 | - | - | - | - | - | - | 0.0692747 |
| **Jefferson St** | 50.5 | 7.5 | - | - | - | - | 49.5 | 10.4 | - | - | 0.06186 |
| **South Dekalb 8/11** | 62.9 | 6.7 | - | - | - | - | 28.8 | 6.3 | 8.3 | 12.7 | 0.0169871 |
| **South Dekalb 2/13** | 42.3 | 13.9 | - | - | - | - | 57.7 | 13.9 | - | - | 0.0792618 |
| **South Dekalb 8/09** | - | - | - | - | 42.5 | 14.5 | 57.5 | 14.5 | - | - | 0.2040939 |
| **Biomass Burning** | - | - | - | - | - | - | - | - | 100.0 | 0.0 | 0.3686555 |
| **CFA** | - | - | - | - | 100.0 | 0.0 | - | - | - | - | 0.1054832 |
| **Diesel** | 70.2 | 11.0 | - | - | - | - | 29.8 | 12.4 | - | - | 0.034851 |

Table S3 Composition of Sulfate in Individual Particles

|  |  |  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Sample ID** | **Ammonium Sulfate** | | **Cu(II) Sulfate** | | **Gypsum** | | **Iron(III) Sulfate** | | **Potassium Sulfate** | | **Jarosite** | | ***R* factor** |
| % | ± | % | ± | % | ± | % | ± | % | ± | % | ± |  |
| **FS8 8.11.11 P1** | 52.7 | 6.5 | 47.3 | 5.0 | - | - | - | - | - | - | - | - | 0.07684 |
| **FS8 8.11.11 P2** | 100.0 | 0.0 | - | - | - | - | 0.0 | 13.8 | - | - | - | - | 0.84504 |
| **FS8 8.11.11 P3** | 14.1 | 11.6 | 56.6 | 16.9 | 29.3 | 8.3 | - | - | - | - | - | - | 0.1339 |
| **FS8 8.11.11 P4** | 57.6 | 11.9 | - | - | 28.5 | 14.5 | - | - | - | - | 13.9 | 23.0 | 0.12948 |
| **FY 6.10.11 P1** | 59.1 | 10.0 | - | - | 32.2 | 7.6 | - | - | 8.6 | 5.0 | - | - | 0.035425 |
| **FY 6.10.11 P2** | 37.3 | 6.4 | 62.7 | 4.7 | - | - | - | - | - | - | - | - | 0.081002 |
| **FY 6.10.11 P3** | - | - | 100.0 | 0.0 | - | - | - | - | - | - | - | - | 0.11171 |
| **FY 6.10.11 P4** | 67.6 | 5.9 | - | - | - | - | - | - | 32.4 | 5.9 | - | - | 0.10811 |
| **FY 9.17.08 P1** | 29.5 | 16.6 | 70.5 | 3.6 | - | - | - | - | - | - | - | - | 0.53495 |
| **FY 9.17.08 P2** | 70.0 | 3.4 | - | - | - | - | - | - | 30.0 | 6.8 | - | - | 0.038327 |
| **FY 9.17.08 P3** | 65.5 | 7.9 | - | - | - | - | - | - | 34.5 | 5.1 | - | - | 0.080905 |
| **FY 9.17.08 P4** | 65.3 | 5.6 | - | - | - | - | - | - | 34.7 | 9.3 | - | - | 0.0143 |
| **FY 9.17.08 P5** | 80.1 | 8.9 | - | - | - | - | - | - | 19.9 | 31.0 | - | - | 0.17596 |
| **SD 8.24.11 P1** | - | - | 64.2 | 3.1 | - | - | 35.8 | 17.3 | - | - | - | - | 0.42401 |
| **SD 8.24.11 P2** | 100.0 | 0.0 | - | - | - | - | - | - | - | - | - | - | 0.49603 |
| **SD 8.24.11 P3** | - | - | - | - | - | - | 100.0 | 16.0 | - | - | - | - | 0.09181 |
| **SD 8.24.11 P4** | 100.0 | 19.4 | - | - | - | - | - | - | - | - | - | - | 0.67987 |