**Supplemental Information**

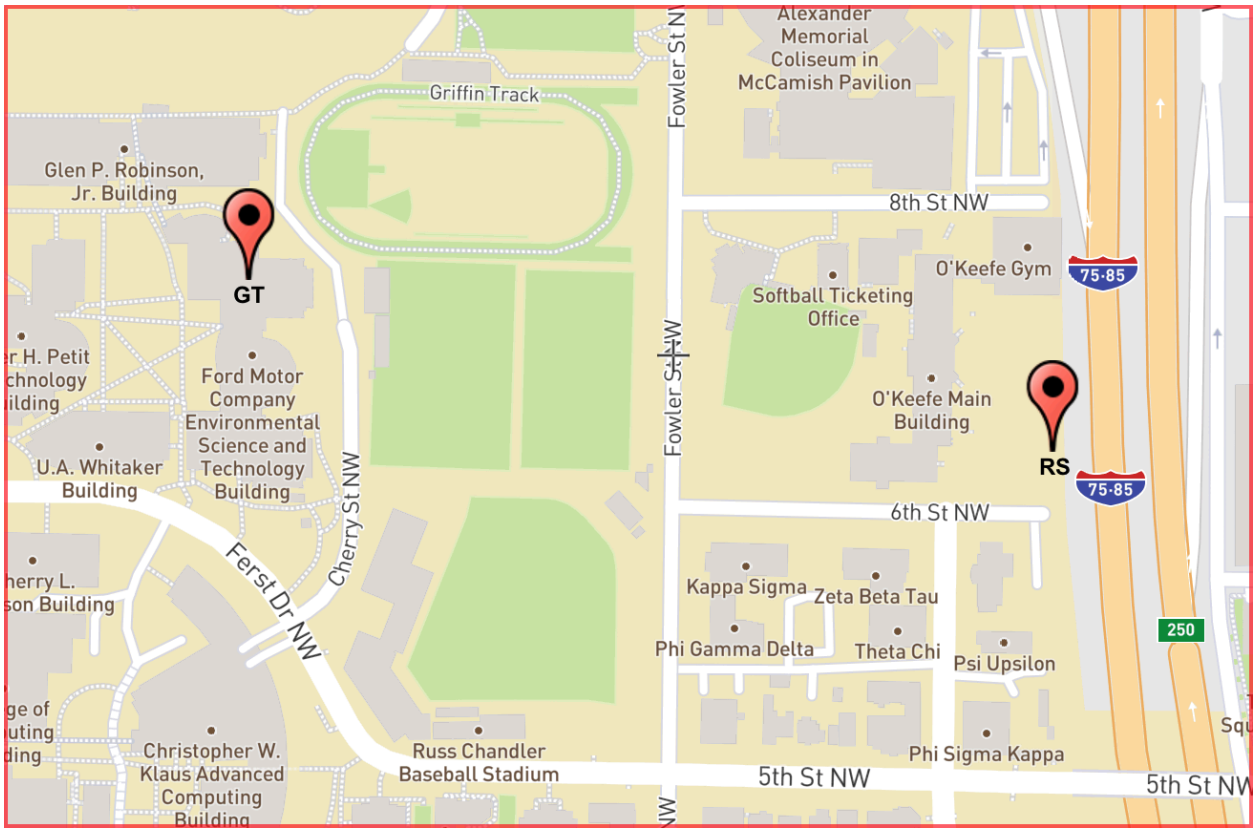


**Figure S1.** Map of sampling sites (Scale is 1:5000). (Map data ©2016 Google Imagery ©2016, DigitalGlobe, Sanborn, U.S. Geological Survey, USDA Farm Service Agency.)

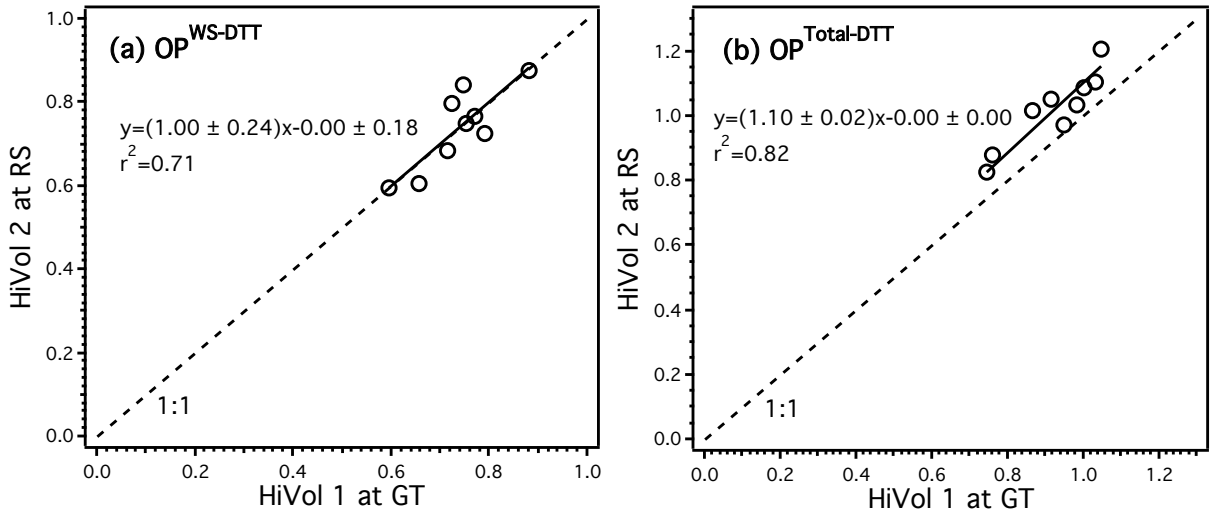


**Figure S2.** (a) $OP^{WS\text{-}DTT}$ (N=9) and (b) $OP^{Total\text{-}DTT}$ (N=9) comparisons for PM samples collected simultaneously at GT using two HiVol sampler. Regression analysis was done by orthogonal regression. The dotted line is 1:1.

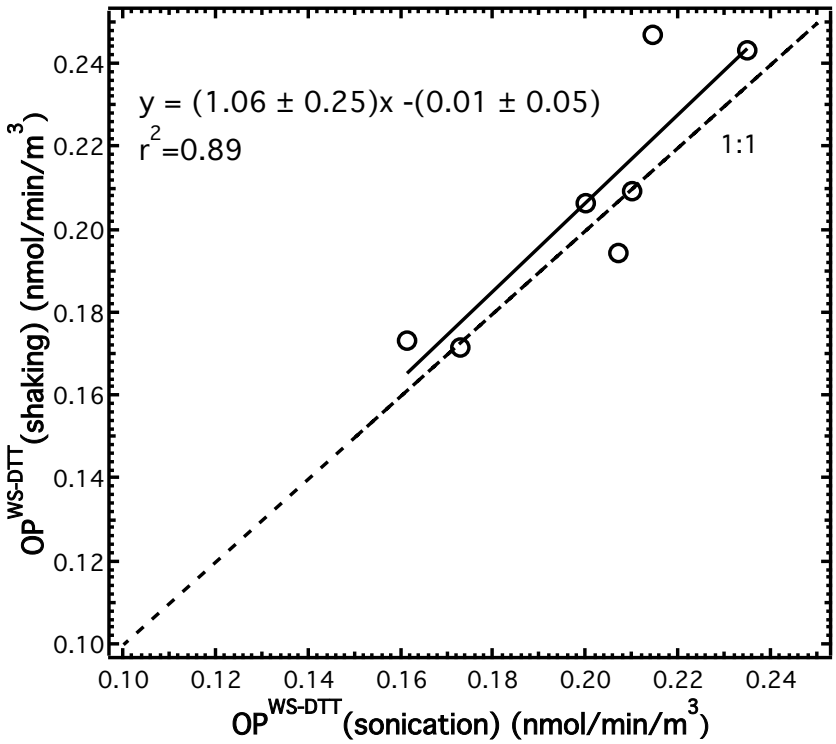


**Figure S3.** 30-minute sonication vs. 2.5-hour shaking comparison for $OP^{WS\text{-}DTT}$ measurements (N=7). Regression analysis was done by orthogonal regression. The dotted line is 1:1.

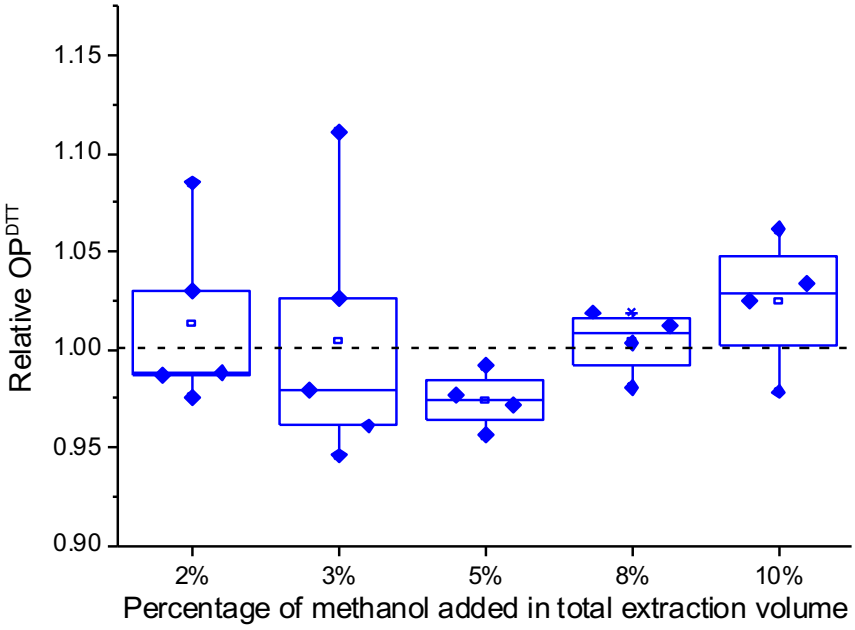


**Figure S4.** The relative $OP^{DTT}$ response (the ratio of $OP^{DTT}$ extracted by methanol-containing solvent to $OP^{DTT}$ extracted by DI only) to adding small amount of methanol into extraction solvent.

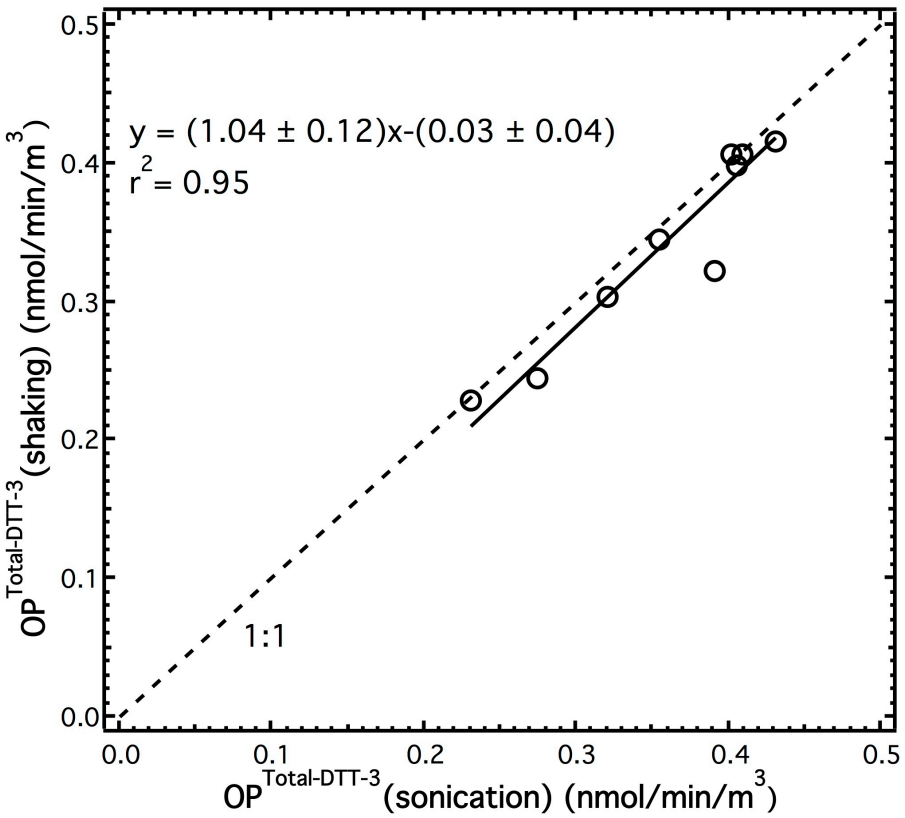


**Figure S5.** 30-minute sonication vs. 2.5-hour shaking comparison for $OP^{Total\text{-}DTT\text{-}3}$ measurements (N=9). Regression analysis was done by orthogonal regression. The dotted line is 1:1.

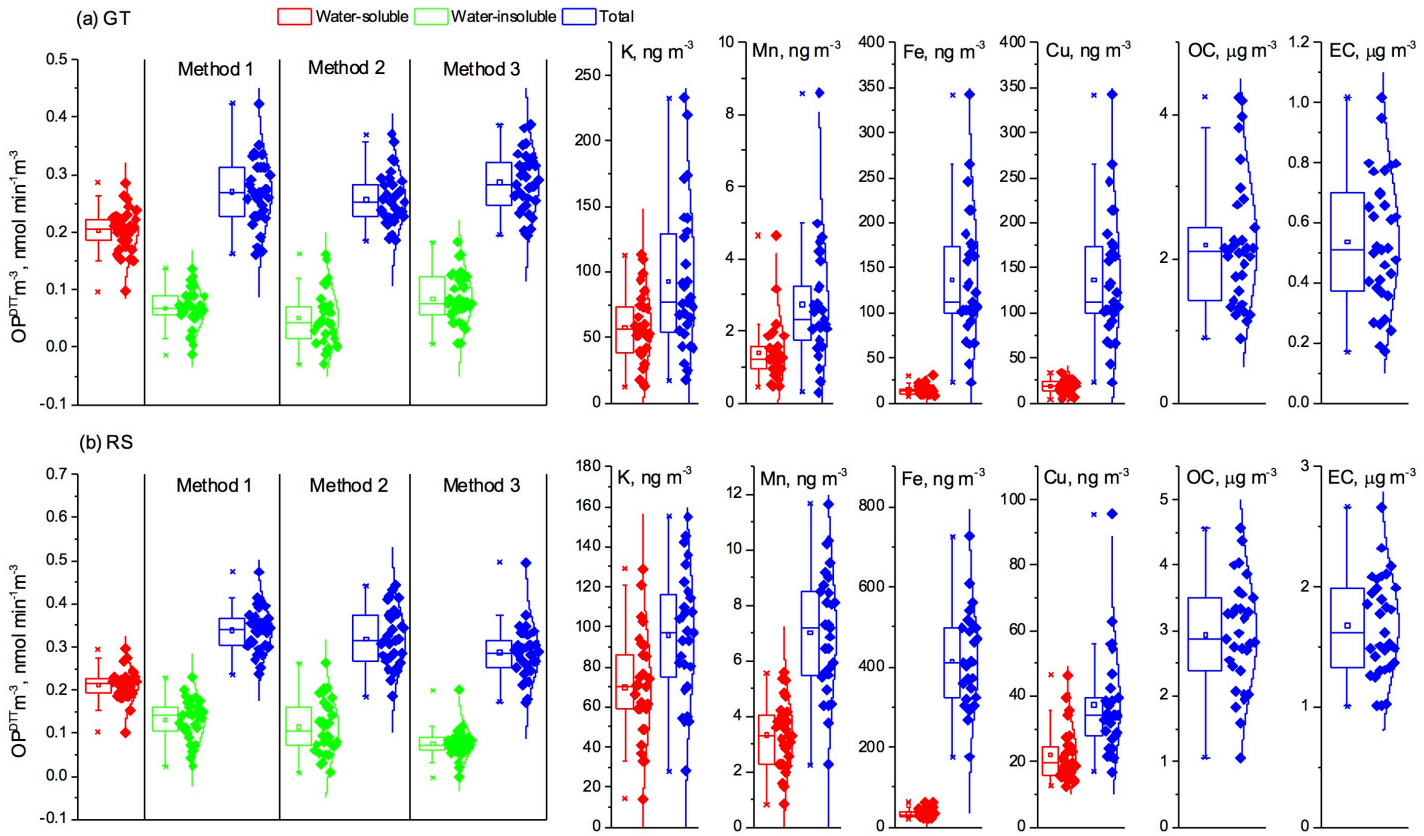


**Figure S6.** Graphical assessment of data normality.

**Figure S1.** Correlation matrix for the various metals and $OP^{DTT}$ obtained by method 3

| **GT** | | | | Water-soluble | | | | | Total | | | | | Water-insoluble | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **N=35** | | OC | EC | DTT | K | Mn | Fe | Cu | DTT | K | Mn | Fe | Cu | DTT | K | Mn | Fe | Cu |
| | EC | **0.83**** | **1** | | | | | | | | | | | | | | | |
| Water-soluble | DTT | **0.79**** | **0.84**** | **1** | | | | | | | | | | | | | | |
| | K | 0.6** | **0.88**** | 0.63** | **1** | | | | | | | | | | | | | |
| | Mn | **0.86**** | **0.7**** | 0.46* | 0.54** | **1** | | | | | | | | | | | | |
| | Fe | **0.8**** | **0.82**** | 0.49* | 0.62** | **0.82**** | **1** | | | | | | | | | | | |
| | Cu | **0.7**** | **0.78**** | **0.77**** | **0.71**** | **0.72**** | 0.64** | **1** | | | | | | | | | | |
| Total | DTT | 0.66** | **0.78**** | **0.71**** | **0.82**** | 0.69** | 0.48* | **0.76**** | **1** | | | | | | | | | |
| | K | 0.67** | **0.84**** | 0.53** | **0.82**** | **0.7**** | 0.59** | 0.52** | 0.69** | **1** | | | | | | | | |
| | Mn | 0.67** | 0.66** | 0.43* | 0.51** | **0.94**** | 0.61** | 0.65** | **0.73**** | **0.7**** | **1** | | | | | | | |
| | Fe | 0.53** | 0.58** | 0.36 | 0.51** | **0.86**** | 0.6** | 0.62** | **0.71**** | 0.56** | **0.97**** | **1** | | | | | | |
| | Cu | **0.72**** | **0.78**** | **0.78**** | 0.62** | **0.87**** | 0.54** | **0.97**** | **0.78**** | **0.7**** | **0.88**** | **0.84**** | **1** | | | | | |
| Water-insoluble | DTT | 0.44* | 0.48** | -0.23 | 0.55** | 0.57** | 0.26 | 0.38 | **0.87**** | 0.50* | 0.66** | 0.63** | 0.56** | **1** | | | | |
| | K | 0.6** | 0.6** | 0.42* | 0.61** | **0.76**** | 0.54** | 0.3 | 0.66** | **0.94**** | **0.74**** | 0.69** | 0.61** | 0.50* | **1** | | | |
| | Mn | 0.43* | 0.66** | 0.31 | 0.37 | **0.79**** | 0.64** | 0.43* | 0.62** | 0.62** | **0.96**** | **0.95**** | **0.75**** | 0.59** | **0.7**** | **1** | | |
| | Fe | 0.49* | 0.57** | 0.35* | 0.49* | **0.86**** | 0.57** | 0.59** | **0.7**** | 0.55** | **0.96**** | **0.9995**** | **0.84**** | 0.64** | 0.69** | **0.95**** | **1** | |
| | Cu | **0.72**** | **0.78**** | **0.78**** | 0.61** | **0.88**** | 0.54** | **0.97**** | **0.78**** | **0.7**** | **0.89**** | **0.85**** | **0.9999**** | 0.56** | 0.63** | **0.76**** | **0.84**** | **1** |

Note: r>0.70 are bold.

**p<0.01. *p<0.05. Correlation not statistically significant is without superscript.

| **RS** | | | | Water-soluble | | | | | Total | | | | | Water-insoluble | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | OC | EC | DTT | K | Mn | Fe | Cu | DTT | K | Mn | Fe | Cu | DTT | K | Mn | Fe | Cu |
| | EC | **0.75**** | **1** | | | | | | | | | | | | | | | |
| Water-soluble | DTT | **0.83**** | **0.79**** | **1** | | | | | | | | | | | | | | |
| | K | **0.86**** | 0.68** | 0.67** | **1** | | | | | | | | | | | | | |
| | Mn | **0.79**** | 0.63** | 0.43* | 0.56** | **1** | | | | | | | | | | | | |
| | Fe | **0.8**** | **0.8**** | **0.88**** | 0.66** | 0.45* | **1** | | | | | | | | | | | |
| | Cu | 0.64** | **0.78**** | 0.54** | **0.77**** | 0.36 | 0.63** | **1** | | | | | | | | | | |
| Total | DTT | **0.71**** | 0.68** | 0.56** | 0.6** | 0.56** | 0.49* | 0.55** | **1** | | | | | | | | | |
| | K | **0.88**** | **0.71**** | 0.69** | **0.9**** | 0.65** | **0.72**** | 0.51* | 0.67** | **1** | | | | | | | | |
| | Mn | **0.86**** | 0.69** | 0.48* | 0.53** | **0.95**** | 0.56** | 0.56** | 0.66** | 0.61** | **1** | | | | | | | |
| | Fe | **0.79**** | **0.75**** | 0.57** | 0.55** | **0.72**** | **0.72**** | **0.71**** | 0.66** | 0.6** | **0.9**** | **1** | | | | | | |
| | Cu | 0.66** | **0.72**** | 0.4 | **0.78**** | 0.65** | 0.56** | **0.93**** | **0.72**** | 0.57** | **0.79**** | **0.84**** | **1** | | | | | |
| Water-insoluble | DTT | -0.34 | -0.37 | -0.51** | -0.47* | -0.37 | -0.40* | -0.40* | **0.84**** | -0.43* | 0.31 | -0.39* | 0.43* | **1** | | | | |
| | K | 0.6** | 0.47* | 0.37 | 0.4 | 0.56** | **0.73**** | 0.28 | 0.47* | **0.7**** | 0.63** | 0.62** | 0.24* | -0.27 | **1** | | | |
| | Mn | **0.77**** | **0.72**** | 0.42* | 0.44* | **0.81**** | 0.62** | 0.65** | 0.67** | 0.49* | **0.95**** | **0.96**** | **0.82**** | 0.31 | 0.57** | **1** | | |
| | Fe | **0.74**** | **0.71**** | 0.47* | 0.49* | **0.71**** | 0.66** | **0.7**** | 0.66** | 0.54** | **0.9**** | **0.998**** | **0.84**** | -0.36 | 0.59** | **0.97**** | **1** | |
| | Cu | 0.67** | **0.73**** | 0.25 | **0.79**** | 0.65** | 0.56** | **0.92**** | **0.73**** | 0.59** | **0.79**** | **0.84**** | **0.9996**** | 0.44* | 0.14 | **0.82**** | **0.84**** | **1** |

Note: r>0.70 are bold.

**p<0.01. *p<0.05. Correlation not statistically significant is without superscript.

N=31 for corrletions between $OP^{DTT}$, N=29 for correlations between $OP^{DTT}$ and PM components.

**Table S2**. $OP^{WS\text{-}DTT}/m^3$ and $OP^{Total\text{-}DTT}/m^3$ (nmol/min/$m^3$)

| Filter type | GT | | | RS | | |
|---|---|---|---|---|---|---|
| | $OP^{WS\text{-}DTT}$ | $OP^{Total\text{-}DTT}$ | $\frac{OP^{WS-DTT}}{OP^{Total-DTT}}$ | $OP^{WS\text{-}DTT}$ | $OP^{Total\text{-}DTT}$ | $\frac{OP^{WS-DTT}}{OP^{Total-DTT}}$ |
| Quartz | $0.20 \pm 0.04$ N=35 | $0.32 \pm 0.06$ N=35 | $65 \pm 10\%$ N=35 | $0.21 \pm 0.03$ N=32 | $0.34 \pm 0.05$ N=33 | $62 \pm 12\%$ N=32 |
| Teflon | $0.13 \pm 0.03$ N=23 | $0.21 \pm 0.04$ N=23 | $65 \pm 14\%$ N=23 | $0.18 \pm 0.02$ N=24 | $0.31 \pm 0.04$ N=24 | $58 \pm 10\%$ N=24 |

**Table S3.** The OP variance on Teflon versus quartz filters assessed by the F-test in ANOVA.

| | F | $F_{critical}$ for $\alpha = 0.05$ |
|---|---|---|
| GT-$OP^{WS\text{-}DTT}$ | 2.082 | 4.013 |
| RS-$OP^{WS\text{-}DTT}$ | 0.499 | 4.020 |
| GT-$OP^{Total\text{-}DTT}$ | 2.084 | 4.013 |
| RS-$OP^{Total\text{-}DTT}$ | 0.159 | 4.016 |

*Null hypothesis assumes that there is no significant difference between Teflon and quartz filters.
*$F < F_{critical}$ when the null hypothesis is true with significance level of 0.05.

**Table S4.** Summary of concentrations of measured PM components.

| | Water-soluble | | Total | |
|---|---|---|---|---|
| | GT | RS | GT | RS |
| K, ng/$m^3$ | 57.49±28.08 N=28 | 69.79±26.56 N=29 | 92.54±56.05 N=28 | 95.97±31.63 N=28 |
| Mn, ng/$m^3$ | 1.40±0.85 N=29 | 3.32±1.20 N=29 | 2.73±1.64 N=29 | 7.02±2.21 N=29 |
| Fe, ng/$m^3$ | 13.94±5.04 N=29 | 33.25±11.36 N=29 | 136.69±70.72 N=29 | 414.41±116.54 N=29 |
| Cu, ng/$m^3$ | 18.17±7.27 N=28 | 22.03±8.29 N=28 | 35.23±18.40 N=29 | 37.20±15.94 N=29 |
| OC, μg/$m^3$ | - | - | 2.19±0.90 N=33 | 2.92±0.82 N=31 |
| EC, μg/$m^3$ | - | - | 0.54±0.22 N=32 | 1.68±0.41 N=31 |

**Table S5.** Coefficients of divergence (CODs) for the paired GT-RS site.

| | Quartz filters | | | | Teflon filters | |
|---|---|---|---|---|---|---|
| | EC | OC | $OP^{WS\text{-}DTT}$ | $OP^{Total\text{-}DTT}$ | $OP^{WS\text{-}DTT}$ | $OP^{Total\text{-}DTT}$ |
| Coefficients of divergence (CODs) | 0.52 | 0.18 | 0.06 | 0.08 | 0.19 | 0.23 |