

Table S1. Weekly averages (\pm standard deviations) of the PM mass and of major chemical components for 3 different size fractions: quasi-ultrafine (quasi-UF), accumulation (ACC), and coarse particles at each site.

| | | unit: $\mu\text{g}/\text{m}^3$ | | | | | | | | | |
|----------|--------|--------------------------------|-----------------|-----------------|-----------------|------------------------------|-------------------------------|-------------------------------|-----------------|------------------------------|-----------------|
| | | PM | OC | EC | Cl ⁻ | NO ₃ ⁻ | PO ₄ ³⁻ | SO ₄ ²⁻ | Na ⁺ | NH ₄ ⁺ | K ⁺ |
| Quasi-UF | SITE 1 | 6.8 \pm 2.59 | 1.84 \pm 0.12 | 0.74 \pm 0.30 | 0.02 \pm 0.01 | 0.13 \pm 0.01 | 0.02 \pm 0.00 | 1.04 \pm 0.29 | 0.03 \pm 0.02 | 0.43 \pm 0.11 | 0.02 \pm 0.01 |
| | SITE 2 | 9.91 \pm 3.09 | 2.45 \pm 0.81 | 0.75 \pm 0.17 | 0.06 \pm 0.03 | 0.23 \pm 0.07 | 0.06 \pm 0.04 | 0.83 \pm 0.25 | 0.05 \pm 0.01 | 0.36 \pm 0.12 | 0.01 \pm 0.01 |
| | SITE 3 | 9.46 \pm 2.48 | 1.82 \pm 0.52 | 0.68 \pm 0.20 | 0.05 \pm 0.06 | 0.20 \pm 0.10 | 0.07 \pm 0.06 | 0.93 \pm 0.20 | 0.08 \pm 0.08 | 0.38 \pm 0.09 | 0.02 \pm 0.01 |
| | SITE 4 | 5.36 \pm 1.70 | 1.91 \pm 1.05 | 0.39 \pm 0.14 | 0.04 \pm 0.02 | 0.31 \pm 0.10 | 0.02 \pm 0.01 | 0.78 \pm 0.33 | 0.04 \pm 0.02 | 0.35 \pm 0.17 | 0.01 \pm 0.01 |
| | SITE 5 | 4.77 \pm 1.86 | 1.28 \pm 0.60 | 0.33 \pm 0.14 | 0.06 \pm 0.11 | 0.12 \pm 0.08 | ND | 0.83 \pm 0.22 | 0.11 \pm 0.19 | 0.28 \pm 0.12 | 0.00 \pm 0.00 |
| | SITE 6 | 7.31 \pm 1.95 | 1.75 \pm 0.37 | 0.65 \pm 0.19 | 0.03 \pm 0.02 | 0.34 \pm 0.17 | 0.03 \pm 0.02 | 0.79 \pm 0.32 | 0.08 \pm 0.11 | 0.38 \pm 0.16 | 0.02 \pm 0.01 |
| ACC | SITE 1 | 4.88 \pm 0.88 | 0.75 \pm 0.23 | 0.16 \pm 0.14 | 0.08 \pm 0.05 | 0.70 \pm 0.26 | 0.08 \pm | 1.62 \pm 0.63 | 0.35 \pm 0.13 | 0.50 \pm 0.30 | 0.03 \pm 0.01 |
| | SITE 2 | 5.87 \pm 1.05 | 0.95 \pm 0.35 | 0.12 \pm 0.09 | 0.29 \pm 0.19 | 1.08 \pm 0.63 | 0.05 \pm 0.01 | 1.89 \pm 1.43 | 0.57 \pm 0.21 | 0.63 \pm 0.60 | 0.04 \pm 0.03 |
| | SITE 3 | 7.34 \pm 3.34 | 0.93 \pm 0.36 | 0.16 \pm 0.09 | 0.21 \pm 0.19 | 0.93 \pm 0.52 | 0.04 \pm 0.02 | 1.73 \pm 1.69 | 0.43 \pm 0.19 | 0.60 \pm 0.66 | 0.03 \pm 0.02 |
| | SITE 4 | 6.92 \pm 2.39 | 1.10 \pm 0.44 | 0.12 \pm 0.07 | 0.22 \pm 0.17 | 1.60 \pm 1.27 | 0.03 \pm 0.03 | 2.23 \pm 2.14 | 0.60 \pm 0.35 | 0.81 \pm 0.99 | 0.03 \pm 0.02 |
| | SITE 5 | 6.15 \pm 1.87 | 0.62 \pm 0.29 | 0.07 \pm 0.04 | 0.27 \pm 0.20 | 0.68 \pm 0.25 | 0.04 \pm 0.03 | 1.03 \pm 0.53 | 0.53 \pm 0.20 | 0.28 \pm 0.17 | 0.02 \pm 0.01 |
| | SITE 6 | 9.32 \pm 4.23 | 1.17 \pm 0.38 | 0.18 \pm 0.14 | 0.20 \pm 0.14 | 2.39 \pm 2.43 | 0.05 \pm 0.03 | 2.25 \pm 2.04 | 0.41 \pm 0.31 | 1.12 \pm 1.05 | 0.03 \pm 0.02 |
| Coarse | SITE 1 | 2.16 \pm 0.34 | 0.28 \pm 0.20 | 0.04 \pm 0.03 | 0.24 \pm 0.12 | 0.50 \pm 0.27 | 0.05 \pm 0.04 | 0.19 \pm 0.05 | 0.29 \pm 0.13 | 0.03 \pm 0.01 | 0.01 \pm 0.01 |
| | SITE 2 | 10.35 \pm 2.82 | 0.90 \pm 0.30 | 0.11 \pm 0.04 | 1.05 \pm 0.52 | 1.48 \pm 0.55 | 0.06 \pm 0.01 | 0.43 \pm 0.13 | 1.00 \pm 0.32 | 0.09 \pm 0.06 | 0.05 \pm 0.01 |
| | SITE 3 | 10.00 \pm 2.32 | 0.75 \pm 0.26 | 0.11 \pm 0.04 | 0.81 \pm 0.50 | 1.41 \pm 0.71 | 0.04 \pm 0.02 | 0.55 \pm 0.13 | 1.21 \pm 1.12 | 0.07 \pm 0.05 | 0.11 \pm 0.20 |
| | SITE 4 | 12.54 \pm 5.94 | 0.95 \pm 0.14 | 0.16 \pm 0.03 | 1.74 \pm 1.13 | 2.04 \pm 1.33 | 0.05 \pm 0.03 | 0.66 \pm 0.30 | 1.50 \pm 0.72 | 0.19 \pm 0.17 | 0.06 \pm 0.03 |
| | SITE 5 | 7.06 \pm 0.75 | 0.39 \pm 0.15 | 0.07 \pm 0.03 | 1.43 \pm 0.88 | 1.26 \pm 0.72 | 0.02 \pm 0.02 | 0.43 \pm 0.18 | 1.20 \pm 0.63 | 0.06 \pm 0.05 | 0.04 \pm 0.02 |
| | SITE 6 | 8.53 \pm 3.60 | 0.73 \pm 0.18 | 0.05 \pm 0.04 | 0.75 \pm 0.47 | 1.95 \pm 1.59 | 0.04 \pm 0.02 | 0.49 \pm 0.33 | 0.90 \pm 0.41 | 0.24 \pm 0.30 | 0.04 \pm 0.03 |

Table S2 Pearson correlation coefficients among water-soluble elements.

| | Al | S | V | Cr | Co | Ni | Cu | Zn | Pb | Mo | Sb |
|----|------|------|------|------|------|------|------|------|------|------|----|
| Al | 1 | | | | | | | | | | |
| S | 0.60 | 1 | | | | | | | | | |
| V | 0.62 | 0.65 | 1 | | | | | | | | |
| Cr | 0.79 | 0.65 | 0.67 | 1 | | | | | | | |
| Co | 0.68 | 0.28 | 0.60 | 0.52 | 1 | | | | | | |
| Ni | 0.65 | 0.67 | 0.99 | 0.71 | 0.59 | 1 | | | | | |
| Cu | 0.49 | 0.14 | 0.35 | 0.45 | 0.27 | 0.33 | 1 | | | | |
| Zn | 0.86 | 0.47 | 0.54 | 0.59 | 0.69 | 0.56 | 0.48 | 1 | | | |
| Pb | 0.60 | 0.82 | 0.76 | 0.79 | 0.34 | 0.80 | 0.15 | 0.34 | 1 | | |
| Mo | 0.78 | 0.56 | 0.80 | 0.94 | 0.59 | 0.82 | 0.51 | 0.62 | 0.76 | 1 | |
| Sb | 0.32 | 0.16 | 0.18 | 0.55 | 0.09 | 0.20 | 0.68 | 0.10 | 0.28 | 0.53 | 1 |

Table S3 Pearson correlation coefficients between redox activities of PM (i.e. macrophage ROS and DTT) and selected chemical species. Coefficients for species in the accumulation and coarse modes were mostly below detection limit; thus values are for quasi-UF mode particles only.

| | Species | Macrophage ROS | | DTT | |
|-------------------------------|---------|----------------|-------------|-------------|-------------|
| | | R | p | R | p |
| Water-Soluble Elements | Li | -0.09 | 0.72 | -0.03 | 0.90 |
| | Na | -0.68 | 0.00 | -0.76 | 0.00 |
| | Mg | -0.70 | 0.00 | -0.74 | 0.00 |
| | K | -0.72 | 0.00 | -0.74 | 0.00 |
| | Ca | -0.64 | 0.01 | -0.68 | 0.00 |
| | Ti | -0.23 | 0.37 | -0.25 | 0.33 |
| | Mn | -0.43 | 0.09 | -0.34 | 0.19 |
| | Fe | 0.13 | 0.63 | 0.18 | 0.48 |
| | Cu | 0.42 | 0.09 | 0.32 | 0.21 |
| | Ba | -0.54 | 0.02 | -0.51 | 0.04 |
| | P | -0.17 | 0.53 | -0.26 | 0.31 |
| | Rb | -0.47 | 0.06 | -0.43 | 0.08 |
| | Sr | -0.76 | 0.00 | -0.80 | 0.00 |
| | Cs | -0.02 | 0.95 | 0.06 | 0.82 |
| | La | -0.04 | 0.88 | 0.03 | 0.90 |
| | Ce | 0.01 | 0.96 | 0.06 | 0.83 |
| | Cd | 0.38 | 0.13 | 0.28 | 0.27 |
| Total Elements | Li | -0.34 | 0.18 | -0.21 | 0.41 |
| | Na | -0.49 | 0.05 | -0.40 | 0.11 |
| | Mg | -0.47 | 0.06 | -0.39 | 0.13 |
| | Al | -0.26 | 0.31 | -0.18 | 0.48 |
| | S | 0.15 | 0.57 | 0.07 | 0.80 |
| | K | -0.25 | 0.33 | -0.21 | 0.42 |
| | Ca | -0.21 | 0.42 | -0.21 | 0.41 |
| | Ti | -0.18 | 0.48 | -0.12 | 0.64 |
| | V | 0.81 | 0.00 | 0.60 | 0.01 |
| | Cr | -0.17 | 0.51 | 0.08 | 0.76 |
| | Mn | -0.28 | 0.27 | -0.16 | 0.54 |
| | Fe | -0.29 | 0.26 | -0.20 | 0.45 |
| | Co | -0.12 | 0.66 | 0.07 | 0.80 |
| | Ni | 0.21 | 0.43 | 0.18 | 0.49 |
| | Cu | -0.27 | 0.29 | -0.22 | 0.41 |
| | Zn | 0.06 | 0.83 | -0.02 | 0.96 |
| | Ba | -0.30 | 0.24 | -0.21 | 0.42 |
| Pb | 0.20 | 0.44 | 0.33 | 0.20 | |

Table S4 Summary of slope, intercept and correlation coefficients (R^2) for the regressions between ROS and DTT levels and selected water-soluble elements[#].

| Species | Macrophage ROS | | | DTT Activity | | |
|-------------------|----------------|-----------|-------|--------------|-----------|-------|
| | Slope* | Intercept | R^2 | Slope | Intercept | R^2 |
| OC | 8.53e-3 | 0.05 | 0.65 | 9.6e-4 | 5.7e-3 | 0.83 |
| WIOC | 9.7e-3 | 0.05 | 0.67 | 1.1e-3 | 6.4e-3 | 0.83 |
| ^a WSOC | 0.04 | 0.08 | 0.46 | 0.49 | 9.43e-3 | 0.43 |
| PAH (MW≤228) | 103.1 | 0.06 | 0.55 | 9.01 | 0.01 | 0.62 |
| V | 3.6 | 0.10 | 0.88 | 0.27 | 0.01 | 0.58 |
| Co | 348.7 | 0.04 | 0.31 | 43.3 | 4.0e-3 | 0.44 |
| Al | 1.70 | 0.14 | 0.25 | 0.22 | 0.02 | 0.41 |
| Zn | 1.01 | 0.11 | 0.25 | 0.10 | 0.01 | 0.22 |
| Mo | 126.9 | 0.05 | 0.40 | 12.6 | 7.7e-3 | 0.50 |

*expressed as (μg Zymosan Units / μg Species)

[#] The level of Quasi-UF ROS at SITE 1 was excessively high (outlier) and discarded from the regression analysis

Table S5 “Best-fitting” regression model results for: a) DTT , and: b) Macrophage ROS. Tests were run and confirmed that the 3 major assumptions of linear regression, i.e. normality (the errors should be normally distributed), homogeneity of variance (the error variance should be constant), and linearity (the relationships between the predictors and the outcome variable should be linear), were valid.

a)

| DTT | Partial R ² | Model R ² | C(p) | F Value | Pr > F | Parameter Estimate | Standardized Estimate |
|-----------|------------------------|----------------------|---------|---------|-------------------------|--------------------|-----------------------|
| Intercept | | | | | | 0.034 | 0 |
| OC | <u>0.76</u> | 0.76 | 40.30 | 44.46 | <u><.0001</u> | 0.056 | 0.64 |
| Al | 0.13 | 0.89 | 13.9851 | 15.35 | 0.0018 | 0.00000915 | 0.37 |
| Co | 0.055 | <u>0.95</u> | 4 | 11.99 | 0.0047 | 0.000739 | 0.25 |

b)

| ROS | Partial R ² | Model R ² | C(p) | F Value | Pr > F | Parameter Estimate | Standardized Estimate |
|-----------|------------------------|----------------------|-------|---------|-------------------------|--------------------|-----------------------|
| Intercept | | | | | | 0.332 | |
| V | <u>0.86</u> | 0.86 | 12.59 | 76.09 | <u><.0001</u> | 0.399 | 0.62 |
| OC | <u>0.07</u> | <u>0.93</u> | 3 | 11.59 | 0.0059 | 0.000223 | 0.41 |

Table S6. Selected water-soluble elements.

unit: ng/m³

| | Quasi-UF | | | | | | ACC | | | | | | Coarse | | | | | |
|----|----------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|
| | SITE 1 | SITE 2 | SITE 3 | SITE 4 | SITE 5 | SITE 6 | SITE 1 | SITE 2 | SITE 3 | SITE 4 | SITE 5 | SITE 6 | SITE 1 | SITE 2 | SITE 3 | SITE 4 | SITE 5 | SITE 6 |
| Li | 0.195 | 0.015 | 0.028 | 0.010 | 0.003 | 0.012 | 0.286 | 0.066 | 0.086 | 0.042 | 0.029 | 0.063 | 0.011 | 0.038 | 0.029 | 0.041 | 0.026 | 0.027 |
| Na | 21.689 | 39.885 | 38.987 | 24.409 | 26.931 | 26.033 | 304.216 | 489.098 | 447.223 | 421.736 | 452.073 | 324.548 | 192.413 | 835.746 | 562.638 | 641.009 | 848.963 | 544.940 |
| Mg | 2.887 | 5.504 | 6.026 | 2.588 | 1.786 | 3.558 | 43.675 | 70.787 | 57.022 | 57.615 | 84.375 | 42.949 | 26.025 | 121.537 | 93.677 | 130.680 | 112.276 | 76.588 |
| Al | 3.762 | 0.937 | 8.387 | 2.015 | 1.297 | 1.834 | 2.718 | 2.152 | 2.888 | 2.644 | 1.552 | 4.285 | 0.580 | 0.435 | 0.271 | 0.932 | 0.342 | 0.332 |
| S | 249.255 | 196.246 | 262.568 | 152.407 | 206.276 | 161.791 | 210.850 | 329.437 | 333.250 | 222.562 | 282.530 | 273.713 | 22.726 | 101.424 | 112.436 | 131.348 | 98.929 | 71.730 |
| K | 8.874 | 9.975 | 7.410 | 4.687 | 2.635 | 6.672 | 18.060 | 25.261 | 22.098 | 21.352 | 25.273 | 17.990 | 8.969 | 46.405 | 33.410 | 44.204 | 36.665 | 30.444 |
| Ca | 0.000 | 6.832 | 0.000 | 0.000 | 0.000 | 0.000 | 20.967 | 44.288 | 50.179 | 19.941 | 62.755 | 23.100 | 11.641 | 205.889 | 207.422 | 177.947 | 129.672 | 114.544 |
| Ti | 0.010 | 0.000 | 0.002 | 0.000 | 0.000 | 0.005 | 0.031 | 0.034 | 0.043 | 0.038 | 0.024 | 0.067 | 0.011 | 0.000 | 0.003 | 0.008 | 0.001 | 0.001 |
| V | 5.401 | 3.843 | 5.007 | 2.394 | 4.550 | 1.791 | 1.036 | 1.303 | 1.691 | 0.859 | 1.209 | 0.872 | 0.164 | 0.345 | 0.590 | 0.368 | 0.670 | 0.098 |
| Cr | 0.056 | 0.029 | 0.050 | 0.025 | 0.015 | 0.045 | 0.030 | 0.030 | 0.041 | 0.031 | 0.017 | 0.048 | 0.006 | 0.012 | 0.014 | 0.026 | 0.009 | 0.014 |
| Mn | 0.660 | 0.544 | 0.613 | 0.236 | 0.119 | 0.490 | 1.309 | 0.670 | 0.772 | 0.677 | 0.374 | 0.822 | 0.247 | 1.575 | 1.302 | 1.506 | 0.695 | 1.304 |
| Fe | 3.525 | 1.053 | 4.505 | 1.744 | 1.468 | 2.343 | 5.210 | 5.209 | 7.746 | 6.516 | 3.130 | 11.797 | 0.696 | 0.112 | 0.337 | 1.761 | 0.839 | 0.719 |
| Co | 0.033 | 0.037 | 0.052 | 0.031 | 0.020 | 0.019 | 0.021 | 0.030 | 0.037 | 0.041 | 0.011 | 0.018 | 0.010 | 0.050 | 0.038 | 0.073 | 0.013 | 0.024 |
| Ni | 1.564 | 1.010 | 1.389 | 0.685 | 1.197 | 0.574 | 0.517 | 0.417 | 0.465 | 0.395 | 0.439 | 0.361 | 0.080 | 0.225 | 0.244 | 0.292 | 0.177 | 0.119 |
| Cu | 1.027 | 1.475 | 2.086 | 0.826 | 1.268 | 1.873 | 0.897 | 1.278 | 1.687 | 2.121 | 0.517 | 2.986 | 0.641 | 0.762 | 1.025 | 2.232 | 1.134 | 2.910 |
| Zn | 5.646 | 9.171 | 15.391 | 1.925 | 2.971 | 3.265 | 6.881 | 6.662 | 5.527 | 6.578 | 3.859 | 6.717 | 2.079 | 4.307 | 4.253 | 6.517 | 3.458 | 4.869 |
| Ba | 0.786 | 0.777 | 0.964 | 0.691 | 0.261 | 1.427 | 2.395 | 1.732 | 2.627 | 4.282 | 1.077 | 5.375 | 1.131 | 2.075 | 2.470 | 5.195 | 1.404 | 6.093 |
| Pb | 0.515 | 0.140 | 0.269 | 0.281 | 0.243 | 0.186 | 0.219 | 0.367 | 0.258 | 0.178 | 0.223 | 0.324 | 0.031 | 0.006 | 0.014 | 0.016 | 0.019 | 0.020 |
| P | 0.380 | 2.098 | 1.784 | 0.000 | 0.000 | 1.105 | 0.377 | 1.473 | 1.334 | 1.002 | 1.071 | 1.431 | 1.276 | 3.250 | 2.171 | 2.419 | 1.997 | 1.960 |
| Rb | 0.010 | 0.012 | 0.010 | 0.006 | 0.004 | 0.008 | 0.019 | 0.019 | 0.019 | 0.017 | 0.019 | 0.015 | 0.006 | 0.025 | 0.019 | 0.028 | 0.017 | 0.017 |
| Sr | 0.000 | 0.059 | 0.118 | 0.000 | 0.000 | 0.135 | 0.458 | 0.635 | 0.708 | 0.636 | 0.681 | 0.642 | 0.208 | 1.330 | 1.815 | 1.990 | 1.301 | 1.308 |
| Mo | 0.165 | 0.091 | 0.150 | 0.073 | 0.060 | 0.119 | 0.064 | 0.069 | 0.090 | 0.069 | 0.043 | 0.116 | 0.018 | 0.055 | 0.056 | 0.090 | 0.034 | 0.085 |
| Cs | 0.001 | 0.001 | 0.001 | 0.001 | 0.001 | 0.001 | 0.002 | 0.003 | 0.003 | 0.004 | 0.004 | 0.003 | 0.000 | 0.000 | 0.000 | 0.001 | 0.000 | 0.000 |
| La | 0.003 | 0.001 | 0.003 | 0.002 | 0.001 | 0.001 | 0.006 | 0.007 | 0.008 | 0.006 | 0.006 | 0.008 | 0.001 | 0.001 | 0.001 | 0.003 | 0.002 | 0.001 |
| Ce | 0.002 | 0.001 | 0.003 | 0.001 | 0.001 | 0.001 | 0.005 | 0.005 | 0.006 | 0.004 | 0.004 | 0.006 | 0.001 | 0.001 | 0.001 | 0.002 | 0.002 | 0.001 |

Figure S1 Sampling sites locations.

Figure S2 Correlation between Macrophage ROS and DTT activities.

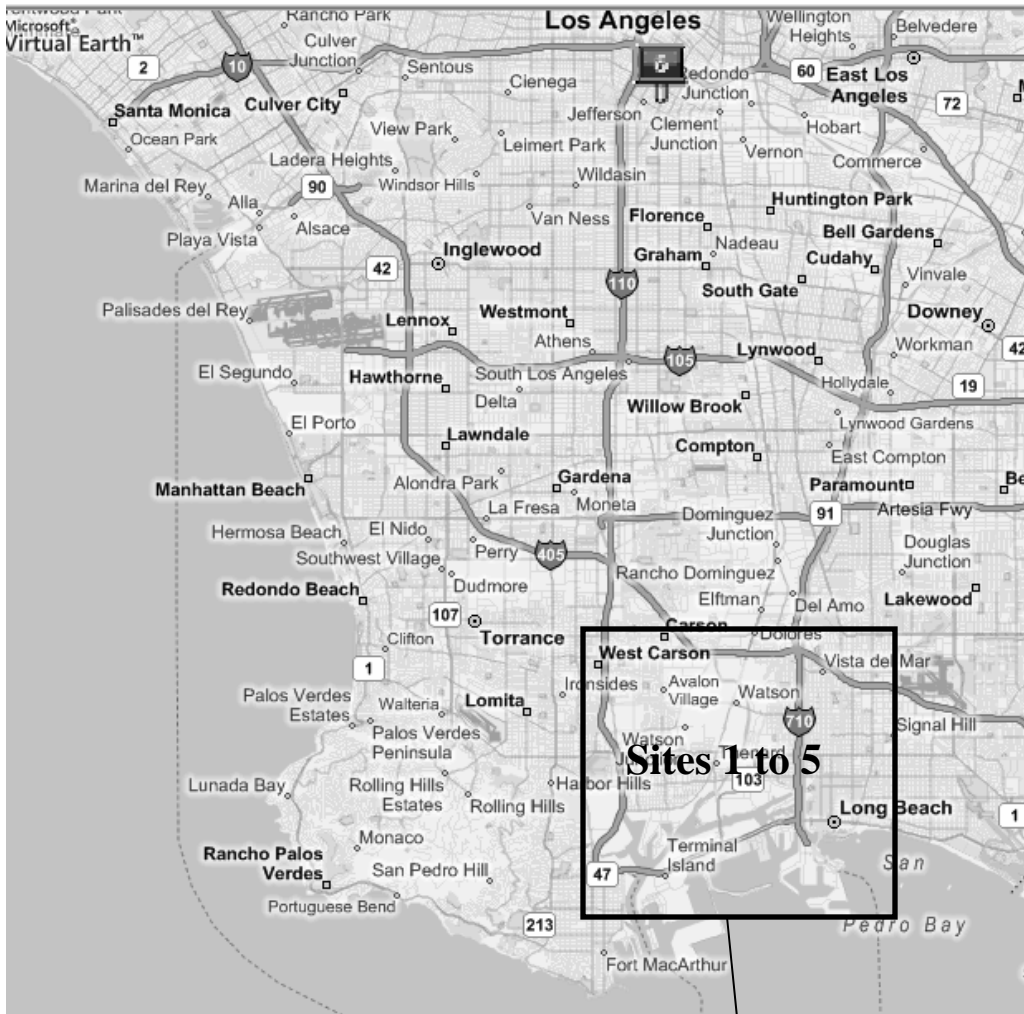


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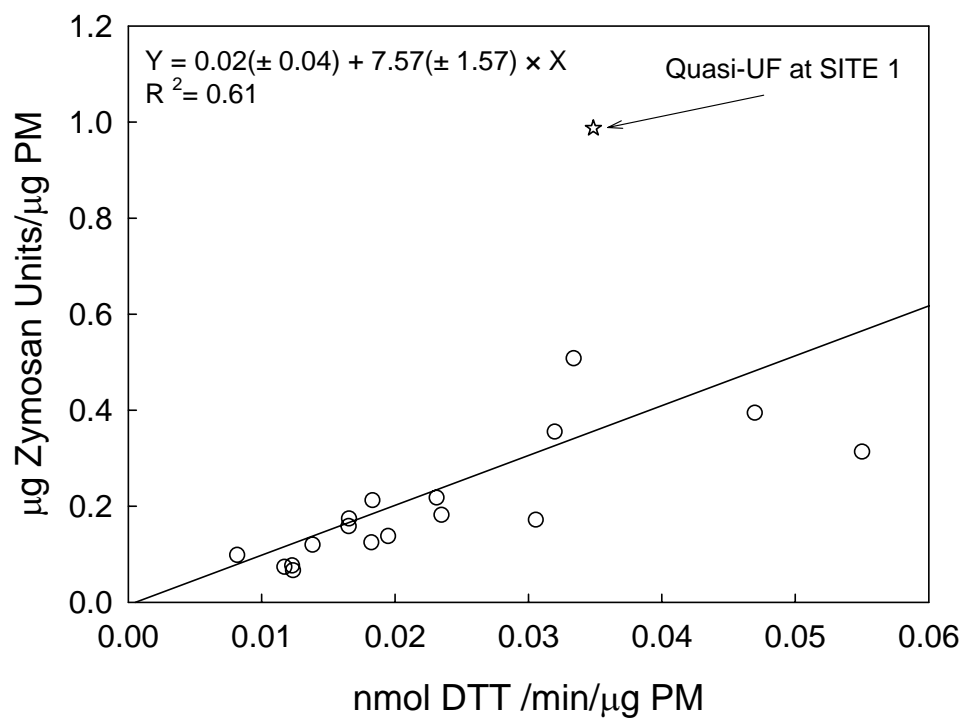


Figure S2 Correlation between Macrophage ROS and DTT activities.