Mass Measurement Method

Triplicate samplers were used indoors and outdoors in all but 4 of the 18 sampling events (in 2 sampling events 2 samplers were used indoors and outdoors, and in 2 other sampling events 1 sampler was used indoors and outdoors). In all (14 x 2 x 3) + (2 x 2 x 2) + (2 x 2 x 1) = 96 sampling filters were used of which 72 filters were weighed against a standard mass using the method described in Section 2.1. The standard mass used was a blank PTFE filter that had been initially weighed in a controlled temperature (21 °C to 22°C) and relative humidity (41 % to 43 %) environment against NIST standard masses (20 mg and 10 mg masses of stainless steel, and 5 mg mass of aluminum) by members of the NIST Mass and Force Group, and its measurement had been corrected for changes in air density due to variations in temperature, pressure and humidity [34]. A correction was made for air buoyancy in the calibration of the standard filter against the metallic standards (0.0122 mg). The buoyancy contribution depends on the product of the air density and the difference in volume between a standard mass and the object being weighed. A blank PTFE filter was chosen as the standard mass (instead of metallic standards) to reduce the buoyancy effect between sampling filters and the standard mass. The buoyancy effect between sampling filters and the standard filter mass was always less than 0.0034 mg (average 0.00067 mg) and was accounted for in the mass measurement calculations.

The mass measurements prior to and after each experiment were made in a room where the temperature was controlled (22.5 °C to 23.5 °C), but the relative humidity was not (it varied between 15 % and 54 % during the measurements over the year). Nonetheless, variability in RH in the weighing room is believed to have had a minimal effect on the mass measurement for the

following reasons: (1) the sampling filter and the standard mass filter were stored in a desiccator for at least 24 hours prior to each mass measurement and they were weighed immediately after removal from the desiccator so the particles on the sampling filter would have minimally absorbed moisture from the air, and (2) each mass measurement of a sampling filter was adjusted with the corresponding mass measurement of the standard filter mass to correct for drifts in weigh scale readings – since the volume of the sampling filters and the standard filter was nearly equal, the buoyancy contribution due to any changes in air density (due to RH, temperature and pressure) would be negligible.

EPR Measurements

The direct measurement of $^\bullet$OH radicals is difficult, and stabilizing $^\bullet$OH with DMPO enables the quantification of the $^\bullet$OH concentration in a sample. However, the DMPO-OH adduct itself can be unstable over time, as reported in a study using γ-irradiation to generate radicals [35]. Based on their measurements, DMPO-OH had a half-life of 80 min when the initial amount of DMPO-OH adduct was on the order of 10^{-7} M and was exposed to an irradiation dose of 10 Gy (they found that the decay rate increases with the irradiation dose). They suggest that homo-dimerization reactions (DMPO-OH/DMPO-OH) and cross-dimerization of DMPO adducts (such as DMPO-OH/DMPO-H) leads to the decay of DMPO-OH over time. In order to minimize an underestimation of the concentration of DMPO-OH in our samples, the time lag between the formation of DMPO-OH adducts and the analysis in the EPR spectrometer was kept to less than 40 minutes. However, no drop in DMPO-OH concentration was observed for a few samples that were reserved for a few hours and re-analyzed. Decay of DMPO-OH may occur over longer time scales, but the rate of decay when DMPO-OH is not exposed to γ-irradiation (as was done in Yoo

et al) remains unknown. The concentration of DMPO-OH in our samples was 1 µmol/L to 10 µmol/L, which was also one to two orders of magnitude higher than the concentrations of DMPO-OH in the samples used in Yoo et al.

Particle Mass Concentration

Indoor and outdoor particle counts were measured in about 5 of the 18 sampling events using an optical particle counter (either CI-7300, Climet; Particle Scan Pro, IQAir; or Aerotrak, TSI). The CI-7300 and Particle Scan Pro instruments were colocated indoors and outdoors a few times, from which a calibration coefficient was estimated for each particle size bin and applied to the data collected. The particle counts from the optical particle counters (OPCs) were used to estimate the mass concentration of particles using the midpoint of every size bin as the representative particle diameter for that size bin, and a particle density of 1.3 g/cm^3 (further details are provided below). This estimate of the mass concentration was multiplied by the sampled volume of air to calculate the mass of particles that would theoretically have been collected on the filters. While the calculated mass was on average two times higher than the actual mass collected on the filters (likely due to inefficient particle samplers, as further discussed below), the calculated mass for outdoor samples was significantly correlated with the actual mass collected (pairwise correlation coefficient 0.96, $p<0.0001$). However, the calculated mass for indoor samples was not significantly correlated with the actual mass collected, which was likely influenced by low indoor particle concentrations. This points to the importance of accurate gravimetric measurements of sampling filters, and is the reason why the analysis regarding particle concentrations and OP^{EPR} on a mass basis relied more heavily on the gravimetric measurements of the particles collected on the sampling filters.

The particle counts from the optical particle counters (OPCs) were used to estimate the mass concentration of particles using basic principles and the assumption that each particle is roughly spherical in shape. Estimates of the density of particles have been reported in published literature using different methods. The density of ambient particles varies with the source of particles, and it can also vary with the size of the particles [36, 37]. Effective densities lower than the density of water (even as low as 0.27 g/cm^3) have been reported for particles composed of lubricating oil and elemental and organic carbon in California [36], whereas coarse mode particles collected during wintertime in Beijing have been reported to have effective densities above 2.0 g/cm^3 [37]. In the absence of more information regarding the chemical composition of particles collected at the research house, an average particle density of 1.3 g/cm^3 was used to convert the number concentration of particles measured in the experiments to mass concentration of particles.

The conversion calculations used (1) the midpoint of every size bin as the representative particle diameter of particles in that bin, and (2) an average particle size of 12 µm for the largest bin in OPCs where the largest bin was for particles >10 µm (CI-7300 and Aerotrak), and an average particle size of 7 µm for the largest bin in the OPC where the largest bin was for particles >5 µm (Particle Scan Pro).

Table S1 presents the indoor and outdoor particle mass concentration calculated from the OPC particle counts. It also provides an estimated theoretical value for the mass of particles that would have been collected over the sampling duration (found by multiplying the total mass concentration estimate by the volume of air sampled). It also presents the ratio of this theoretical

mass to the actual mass of particles collected on the sampling filters. The theoretical masses were on average 2.3 times higher than the actual mass of particles collected on the filters. The ratio of theoretical to actual mass of particles collected was under 3.2 for all sampling events except for one outdoor sample (sampling event ending on 2-5-16), which had a theoretical mass 7.6 times the actual mass of particles collected. Some differences were expected in the local particle concentration in the bedroom (where the OPC was located) and in the dining room (where the sampling filters were located), and the conversion factors for colocated instruments and uncertainty in the assumed particle density in the mass calculations would also play a role in differences between the theoretical and actual mass of particles collected. Nonetheless, the theoretical masses are also higher because the TSP samplers did not efficiently collect all airborne particles. This became evident when a few experiments were conducted at the end of the sampling campaign to collect $PM_{2.5}$ using personal environmental monitors, and it was found that the mass of particles collected with the TSP samplers was similar to the mass of particles collected with the $PM_{2.5}$ samplers. Some particles were likely passing through the filter in the TSP samplers. This implies that on a volume basis the OP^{EPR} of particles in this study should be higher than what is reported (since not all particles in the air flow were collected on the filters). However, on a mass basis the OP^{EPR} of particles should give a fairly correct representation because the oxidative potential of only those particles was measured that were actually collected on the filters (particles not collected on the filters obviously did not get included in the mass and OP^{EPR} measurement).

Higher mass concentrations of particles >1 µm were present when the relative humidity was high indoors. However, the mass of particles collected on the sampling filters was not as high as the

indoor particle mass concentrations would indicate (Table S1). This is likely due to the heavy water content of the indoor aerosols generated when the humidifiers were being run, and the water got evaporated from the filters when they were conditioned in the desiccator before weighing.

The cooking tests (especially the one in which the fire alarm went off) resulted in some of the highest masses of particles collected on the sampling filters. The optical particle counter was located in the front bedroom (to facilitate the use of a switching device to use the same instrument for indoor and outdoor sampling), whereas the sampling filters were set up in the dining room, a central area of the house about 4 m from the kitchen stove. Many of the particles generated during cooking would have gotten deposited onto various indoor surfaces and walls before reaching the room with the OPC, which is why the data from the OPC during the cooking test was not higher than in the tests when cooking was not conducted.

Air Change Rate at Research House

The forced-air fan was kept on during the 4-day sampling events in this study so that the indoor environment was well-mixed (no additional mixing fans were used to avoid resuspending settled dust). Nabinger and Persily [23] previously characterized λ at the research house before and after retrofits were done to tighten the building envelope and reduce duct leakage. In their studies with the forced-air fan on after the retrofits, Nabinger and Persily measured air change rates as low as about 0.1 /h when the wind speed was less than 2 m/s, or when indoor temperatures were only 0 °C to 10 °C higher than outdoor temperatures. Air change rates as high as about 0.4 /h were recorded when the wind speed was about 8 m/s, or about 0.3/h when the indoor-outdoor

temperature difference increased to 25 ℃. λ measured during the eight sampling events conducted under normal indoor conditions (22 °C to 26 °C) in this study ranged from 0.16 /h to 0.32 /h, with higher values (0.45 /h to 0.58 /h) measured during the sampling events when indoor temperature was elevated to 32 °C to 34 °C. In general, λ measured in this study and its weather dependence is consistent with what Nabinger and Persily observed after the building retrofits.

RH and Temperature at Research House

Indoor RH was elevated during two sampling events using portable humidifiers filled with distilled water. In the first sampling event a single humidifier was used (average indoor RH 32.5 % ± 1.4 %), and in the second event three humidifiers were used which led to a much higher indoor RH level (also due to the outdoor RH being quite high, around 90 %; average indoor RH 62.9 % ± 3.9 %). In the two sampling events when both temperature and RH was increased (using two or three humidifiers), indoor RH remained between 23 % and 29 % as the elevated indoor temperature increased the moisture holding capacity of air. Mass normalized OP^{EPR} of indoor particles was correlated with indoor RH (Pearson's correlation coefficient 0.42, p=0.0108) and inversely correlated with indoor temperature (Spearman's rank correlation coefficient -0.40, p=0.0160). The temperature, relative humidity, and humidity ratio during the sampling events when indoor conditions were modified are presented in Table S2.

OP^{EPR} of Indoor Particles as a Function of Indoor RH

The use of humidifiers increased the mass concentration of particles in the air by increasing the water content of existing particles and likely facilitating chemical reactions which generate particles. OP^{EPR}/mg particles is a measure of OP^{EPR} per dry mass of particles, and it would

overestimate OP^{EPR} per mass of particles collected during the higher RH sampling events, given that the water content of the particles collected on the sampling filters would diminish during the 24-hour conditioning step in the desiccator prior to measuring the post-sampling mass of the filters. Nonetheless, the high value of OP^{EPR}/mg particles indicates that particles generated under high indoor RH conditions at the research house have a higher oxidative potential. This may be due to the desorption of other pollutants from the walls, carpet and other surfaces as water vapor gets adsorbed onto these surfaces. The research house is a manufactured house with building materials that were not selected to be low emitting [38], and it is likely that VOCs, metals, and other chemicals desorb from indoor surfaces and attach onto existing particles or participate in chemical reactions generating particles. However, chemical characterization of indoor particles would have to be conducted to properly identify the sources of these particles.

Cooking at Research House

During two sampling events, residential cooking activities were simulated by heating oil in a frying pan to 150 °C to 180 °C and boiling water in a pot to 85 °C to 90 °C (temperatures were measured with an IR thermometer) for 45 minutes, 3 times per day to simulate breakfast, lunch and dinner. In the first cooking test, 194 µg of particles were collected over the 4-day sampling period, whereas 709 µg of particles were collected during the second cooking test when heating oil for "breakfast" on the second day of the test had generated so much smoke that the fire alarm had been set off.

Normality Tests on Data Distributions and Residuals of Linear Regression Models

Tables S3 and S4 report the particle mass on the sampling filters and the corresponding OP^{EPR} for indoor and outdoor particles collected during the 8 sampling events conducted during normal operating conditions and the 10 sampling events conducted under different indoor conditions at the research house, respectively.

The Shapiro-Wilk test for normality was used to determine if the OP^{EPR} of particles followed a normal distribution. OP^{EPR} was deemed to fit a normal distribution if $p>0.05$. For the eight sampling events which were conducted during normal operating conditions, OP^{EPR} of indoor ($W=0.91$, $p=0.34$) and outdoor ($W=0.92$, $p=0.41$) particles on a volume basis followed a normal distribution, as did OP^{EPR} of indoor ($W=0.91$, $p=0.44$) and outdoor ($W=0.98$, $p=0.92$) particles on a mass basis. This led to the selection of pairwise t-tests (rather than non-parametric equivalents) to compare the means of these data sets.

For the entire data set of 18 experiments, the indoor and outdoor relative humidity, outdoor wind speed and direction, and outdoor temperature followed normal distributions. The air change rate was not normally distributed and followed a lognormal distribution, as expected ($W=0.92$, $p=0.17$ for log-transformed air change rate data). Indoor temperature was regulated by a thermostat and did not follow a normal (or lognormal) distribution. Volume normalized OP^{EPR} of indoor particles, mass normalized OP^{EPR} of outdoor particles, and mass of particles collected on indoor sampling filters were also not normally distributed when the entire data set of 18 experiments was considered. Any correlation test involving the data that was not normally distributed was run with a non-parametric test i.e. Spearman's rank correlation test.

A linear regression model can be developed with variables that do not follow a normal distribution, so non-normality of a few variables did not limit the development of the model for OP^{EPR} of indoor particles. It is necessary, however, for the residuals from a linear regression model to be normally distributed for the t-tests on the coefficients of the predictor variables to be valid. The residuals from the regression on volume normalized OP^{EPR} of indoor particles were checked with the Shapiro-Wilk test, kernel density plot, standardized normal probability (P-P) plot and quantile-quantile (Q-Q) plot, and were seen to approximately follow a normal distribution. Another assumption for linear regression models is that the variance of the residuals is homogeneous. The White's test and the Breusch-Pagan test were used to verify that the residuals were indeed homoscedastic, i.e., the variance in the residuals was homogeneous. Due to the results of these checks on the linear regression model, it was not deemed necessary to transform any of the variables.

A linear regression model was also developed for mass normalized OP^{EPR} of indoor particles. Mass normalized OP^{EPR} of outdoor particles was controlled for in this model, as discussed in the paper. The residuals from this model appeared to be normally distributed (Shapiro-Wilk test $p=0.2559$). The variance of the residuals appeared to be homogeneous (Breusch-Pagan test $p=0.4591$). Since the infiltration of outdoor particles into the indoor space is a function of the volume of outdoor air brought into the building, a variation of the model was developed which controlled for volume normalized OP^{EPR} of outdoor particles. But that model had residuals which were not normally distributed (Shapiro-Wilk test $p=0.0094$) and appeared to be heteroscedastic (Breusch-Pagan test $p=0.0108$), and was, thus, not considered.

In the linear regression model for volume normalized OP^{EPR} of indoor particles, an interaction

term of temperature and RH was considered (because of the interaction between the two) but it

was not seen to be significant in the model and was not included.

Table S1. Indoor and outdoor particle mass concentrations estimated from OPC particle counts.
The estimated particle mass that would have been collected, and the ratio of estimated to actual
particle mass collected on sampling filters is also given. The standard deviation (SD) of
measurements over the 4-day sampling periods is provided.

Sampling Stop Date	Indoor Conditions	Indoor Particle Concentration [µg/m³]								Estimated PM Collected [µg]		Estimated / Actual Mass Collected [-]
		0.3-1 µm	SD	1-5 µm	SD	>5 µm	SD	Total	SD	Ave	SD	
2/5/2016	High T&RH											
4/12/2016	High λ	0.4	0.0	0.1	0.1	0.1	0.2	0.6	0.2	71.9	25.9	0.8
4/22/2016	Cooking	0.7	0.4	0.1	0.2	0.1	0.3	0.8	0.5	98.1	61.3	0.1
5/9/2016	High λ	0.2	0.1	0.4	0.5	0.6	1.4	1.2	1.5	133.7	166.0	1.5
5/13/2016	High RH	0.7	0.3	0.8	1.2	1.1	3.2	2.6	3.4	308.6	400.3	2.0
5/23/2016	Baseline	0.1	0.0	0.2	0.6	0.5	1.8	0.9	1.9	105.1	233.6	1.7
9/14/2016	Baseline	0.7	0.5	0.5	0.6	0.9	1.3	2.1	1.5	278.7	200.8	3.2

Sampling Stop Date	Indoor Conditions	Outdoor Particle Concentration [µg/m³]								Estimated PM Collected [µg]		Estimated / Actual Mass Collected [-]
		0.3-1 µm	SD	1-5 µm	SD	>5 µm	SD	Total	SD	Ave	SD	
2/5/2016	High T&RH	0.8	0.4	6.1	4.7	1.5	2.9	8.4	5.6	958.1	637.8	7.6
4/12/2016	High λ	0.4	0.4	3.5	10.7	0.7	1.7	4.6	10.8	544.8	1274.0	1.9
4/22/2016	Cooking											
5/9/2016	High λ	0.6	0.5	3.8	2.4	1.0	2.5	5.5	3.5	617.6	396.2	2.2
5/13/2016	High RH	3.5	1.7	18.7	12.3	1.4	3.3	23.6	12.8	2744.4	1492.7	3.0
5/23/2016	Baseline	1.0	0.4	5.5	1.9	1.2	1.6	7.7	2.5	918.3	299.8	1.7
9/14/2016	Baseline											

Table S2. Average relative humidity, humidity ratio, and temperature during all the sampling

events. - SD represents standard deviation and uncertainty in the measurements.

Sampling Stop Date	Indoor Conditions	Temperature [°C]				Relative Humidity [%]				Humidity Ratio [g water / kg dry air]	
		Out	SD	In	SD	Out	SD	In	SD	Out	In
11/20/2015	Normal	13.1	3.7	22.1	0.9	73.3	23.6	39.4	2.2	6.9	6.5
3/15/2016	Normal	11.3	3.6	24.5	0.5	76.3	21.0	29.4	1.7	6.3	5.6
3/21/2016	Normal	6.9	4.8	22.3	0.5	58.5	24.3	26.2	1.6	3.6	4.4
4/18/2016	Normal	13.6	6.1	26.4	1.8	48.7	19.3	22.9	1.3	4.7	4.9
5/23/2016	Normal	15.1	4.5	23.9	0.9	80.2	24.0	41.0	2.7	8.6	7.6
9/14/2016	Normal	24.4	4.7	25.9	0.8	68.4	17.1	51.0	2.2	13.1	10.6
1/31/2016	High T	1.5	5.4	33.1	1.1	63.7	16.0	12.2	0.6	2.7	3.8
2/23/2016	High T	7.9	4.1	34.3	4.0	71.2	20.2	15.3	0.7	4.7	5.1
1/14/2016	High RH	-1.6	4.8	20.0	0.6	49.5	10.1	32.5	1.4	1.7	4.7
5/13/2016	High RH	15.4	3.0	24.4	1.1	87.5	13.0	62.9	3.9	9.6	12.0
2/5/2016	High T&RH	6.3	3.2	33.0	0.8	75.3	19.5	23.9	1.3	4.4	7.5
2/29/2016	High T&RH	5.1	6.0	32.3	0.7	48.7	12.0	27.3	1.6	2.6	8.2
4/12/2016	High λ	8.7	6.7	22.0	0.8	55.2	16.9	23.9	1.5	3.8	3.9
5/9/2016	High λ	19.1	9.1	22.0	1.0	80.2	22.3	41.4	3.2	11.1	6.8
4/1/2016	Cooking	13.2	5.6	25.3	0.8	53.4	16.5	30.8	2.2	5.0	6.2
4/22/2016	Cooking	17.4	5.6	24.7	0.8	47.9	19.2	28.5	2.4	5.9	5.5

Table S3. Particle mass on sampling filters and OP^{EPR} of indoor and outdoor particles (volume normalized and mass normalized) collected during eight 4-day sampling events when the research house was operated under normal conditions*.

- SD represents standard deviation of triplicate samples, when applicable.
* Mass of particles was measured in 5 out of the 8 sampling events.

Sampling Stop Date	PM Mass [mg]					OP^{EPR} [µM/100m^3 air]					OP^{EPR} [µM/mg particles]				
	Outdoor	SD	Indoor	SD	In/Out	Outdoor	SD	Indoor	SD	In/Out	Outdoor	SD	Indoor	SD	In/Out
9/4/2015						2.37		0.46		0.19					
9/18/2015						2.94		2.20		0.75					
11/20/2015						1.26	1.05	1.09	0.79	0.87					
3/15/2016	0.498	0.083	0.086	0.012	0.173	4.22	0.27	3.12	0.13	0.74	10.20	1.10	43.86	8.36	4.30
3/21/2016	0.277	0.068	0.078	0.033	0.283	2.14	0.30	2.17	0.63	1.01	9.52	3.22	34.31	7.07	3.60
4/18/2016	0.627	0.096	0.058	0.024	0.092	3.93	0.50	1.78	0.55	0.45	7.18	0.29	23.45	2.48	3.27
5/23/2016	0.552	0.083	0.061	0.026	0.110	1.76	0.59	0.65	0.15	0.37	3.78	0.95	14.67	8.24	3.88
9/14/2016	0.758	0.020	0.088	0.001	0.117	1.48	0.34	0.46	0.25	0.31	2.61	0.65	6.91	3.82	2.65

Table S4. Particle mass on sampling filters and OP^{EPR} of indoor and outdoor particles (volume normalized and mass normalized) collected during ten 4-day sampling events when temperature, relative humidity, air change rate and cooking activities were modulated inside the research house.

- SD represents standard deviation of triplicate samples.
* Mass of particles was measured in 7 out of the 10 sampling events.

Sampling Stop Date	Indoor Conditions	PM Mass [mg]					OP^{EPR} [µM/100m^3 air]					OP^{EPR} [µM/mg particles]				
		Outdoor	SD	Indoor	SD	In/Out	Outdoor	SD	Indoor	SD	In/Out	Outdoor	SD	Indoor	SD	In/Out
1/31/2016	High T	0.470	0.052	0.077	0.008	0.164	3.06	0.08	0.54	0.27	0.18	7.97	1.06	8.30	3.55	1.04
2/23/2016	High T						2.95	1.16	1.09	0.33	0.37					
1/14/2016	High RH						5.07	0.95	1.71	1.40	0.34					
5/13/2016	High RH	0.914	0.055	0.155	0.007	0.170	3.55	0.39	8.21	0.43	2.31	4.56	0.79	61.76	4.54	13.56
2/5/2016	High T&RH						3.73	0.70	4.22	0.90	1.13					
2/29/2016	High T&RH	0.177	0.031	0.173	0.026	0.981	2.50	0.32	4.84	1.29	1.93	17.21	4.86	34.14	13.19	1.98
4/12/2016	High λ	0.290	0.045	0.095	0.018	0.328	1.38	0.46	1.68	0.23	1.22	5.88	2.94	20.95	2.21	3.56
5/9/2016	High λ	0.276	0.053	0.089	0.046	0.323	0.70	0.31	2.25	0.08	3.24	2.75	0.78	35.49	21.11	12.89
4/1/2016	Cooking	0.454	0.076	0.194	0.020	0.427	0.97	0.36	1.27	0.14	1.31	2.59	1.35	7.56	0.23	2.92
4/22/2016	Cooking	0.812	0.106	0.709	0.041	0.873	2.37	0.42	0.96	0.35	0.41	3.48	0.67	1.61	0.59	0.46