Supplemental Material

Can ΔPM2.5/ΔCO and ΔNOy/ΔCO Enhancement Ratios Be Used to Characterize the Influence of Wildfire Smoke in Urban Areas?

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Results:

Comparison of AERs with EPA County ERs

We calculated PM2.5/CO and NOx/CO ERs for each county using the NEI11 from the EPA to compare the PM2.5/CO and NOy/CO AERs (*https://www.epa.gov/air-emissions-inventories/2011-national-emissions-inventory-nei-data*). CO and NOx emissions for all counties are dominated by mobile sources, while the main sources of PM2.5 emissions are more county dependent. We directly compared AERs calculated from ambient hourly data to the corresponding ERs derived from the emissions inventory (Parrish et al., 2002; Parrish, 2006; Parrish et al., 2009; Pollack et al., 2013).

PM2.5/CO AERs and EPA PM2.5/CO ERs

PM2.5/CO ERs were calculated using the sum of all emission sources, the sum of all sources except fires, and the sum of all sources except fires and dust to compare with the PM2.5/CO AERs (Table S2). In addition PM2.5/CO ERs were calculated for fuel combustion sources and mobile sources as they have the highest and lowest ratios, respectively. The PM2.5/CO ER for fuel combustion is very similar in all counties except for those of Boise and Portland. In the emission inventory, Ada County (Boise) and Multnomah County (Portland) are heavily influenced by industrial and commercial biomass combustion (~40% of fuel combustion PM2.5 emissions). Industrial and commercial biomass combustion have a large EPA PM2.5/CO ER (0.854 $\mu g\ m^{-3}\ ppbv^{-1}$), which skews the total fuel combustion PM2.5/CO ER higher. Most other counties' fuel combustion emissions are dominated by residential wood combustion.

For the summed EPA PM2.5/CO ERs,removing fire emissions produces a negligible change at all sites except for Fresno and Chico. Removing the dust emissions in addition to fire emission changes the PM2.5/CO ER by at least 15% at all sites. The most significant differences are seen in Washoe County (Reno), Ada County (Boise), Denver County, and Fresno County. The dust source is primarily from construction and unpaved roads, and therefore may be location dependent within the county. Comparing the PM2.5/CO ERs calculated from the sum of emissions except fires and dust, 8 of the 9 sites are within 30% of the measured PM2.5/CO AERs. Portland is exception, the measured PM2.5/CO AER (0.030 µg m^{-3} $ppbv^{-1}$) is lower than the PM2.5/CO ER for sum except fires and dust (0.050 µg m^{-3} $ppbv^{-1}$). Comparing PM2.5/CO ERs calculated from the sum of emissions except fires, only 5 of the 9 sites are within 30% of the measured PM2.5/CO AERs. The differences between ambient PM2.5/CO AERs and emission inventory ERs may be due to the spatial inhomogeneity of the monitoring sites, the production/loss of PM2.5 after emission, and the lack of spatial and temporal resolution of the emission inventories.

NOy/CO AERs and EPA NOx/CO ERs

Table S3 shows NOy/CO AERs measured at the monitoring sites, as well as NOx/CO ERs calculated from the EPA emission inventories for the sum of all emission sources, the sum of all sources except fires, mobile sources, and fuel combustion sources. NOy is a conserved measure of emitted NOx, so NOy/CO AERs are a more accurate representation of NOx/CO ERs than NOx/CO AERs (Seinfeld and Pandis, 2006). As the principal source of NOx and CO are vehicles, the NOx/CO ERs are dominated by the mobile NOx/CO ER.

The NOx/CO ERs sum of all sources were within 30% of the NOy/CO AERs for 4 of the 6 sites. Portland and Fresno are the exceptions, the NOx/CO ERs being higher than the NOy/CO AERs by a factor of ~2 and 5, respectively. The discrepancy between the AERs and ERs can be due to a number of possibilities such as the lack of spatial and temporal resolution of the emission inventories, or uncertainties in the emissions inventories. Loss of NOy due to deposition between emission and measurement does not appear to be a large influence. As previously mentioned, the high R^2 values for NOy/CO AERs and the lack of significant diurnal variation in NOy/CO suggest a homogenously mixed source that does that substantially vary. One issue that may lead to the discrepancy is that the monitoring sites may be representative of local sources and not the entire county. An example of this is Fresno, which has a large difference between the NOy/CO AER and NOx/CO ER. Fresno County contains a ~70 mile stretch of the I-5 highway that is nearly 50 miles from the city of Fresno. The Fresno County NOx/CO total ER is very high due to heavy diesel vehicles, most likely from the I-5 corridor, which has little influence in the city of Fresno. For the Portland site both the PM2.5/CO ER and NOx/CO ER are significantly higher than the measured PM2.5/CO AER and NOy/CO AER. This may indicate the Portland site is not indicative of the county as a whole.

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Table S1. EPA sample site location and names. The X indicates that data for the parameter was available.

City	County	EPA site name	CO	PM2.5	NOy	# WF events
Seattle, WA	King	Seattle - Beacon Hill	X	X	X	1
Seattle, WA	King	Seattle - 10th St and Weller	X	X		1
Portland, OR	Multnomah	Portland - SE Lafayette	X	X	X	2
Boise, ID	Ada	St Luke's Meridian	X	X	X	6
Denver, CO	Denver	La Casa	X	X	X	1
Stockton, CA	San Joaquin	Stockton-Hazelton	X	X		1
Fresno, CA	Fresno	Fresno - Garland	X	X	X	4
Reno, NV	Washoe	Reno3	X	X	X	5
Chico, CA	Butte	Chico - East Avenue	X	X		4

Table S2.

PM2.5/CO AERs for each site and PM2.5/CO ERs calculated from the county emission inventories. The PM2.5/CO AERs were calculated using an RMA regression of all data up to the 99th percentile of PM2.5 mass. The county ERs were calculated for the mobile sources, fuel combustion sources, the sum of all sources, all sources except fires, and all sources except fires and dust.

		PM2.5/CO AERs (µg m^{-3} ppbv^{-1})*		**NEI11 County PM2.5/CO ERs (µg m^{-3} ppbv^{-1})**				
Site location	**Site county**	**slope**	**R^2**	**Mobile**	**Fuel combustion**	**Sum of all sources**	**Sum except fires**	**Sum except fires and dust**
Seattle - Beacon Hill	King	0.035	0.379	0.010	0.173	0.038	0.038	0.030
Seattle - 10th St	King	0.021	0.407	0.010	0.173	0.038	0.038	0.030
Portland, OR	Multnomah	0.030	0.537	0.010	0.237	0.063	0.062	0.050
Boise, ID	Ada	0.066	0.348	0.010	0.245	0.081	0.080	0.053
Denver, CO	Denver	0.021	0.188	0.008	0.155	0.037	0.037	0.022
Stockton, CA	San Joaquin	0.046	0.351	0.013	0.157	0.050	0.046	0.036
Fresno, CA	Fresno	0.041	0.454	0.014	0.194	0.063	0.057	0.041
Reno, NV	Washoe	0.029	0.315	0.009	0.156	0.066	0.062	0.027
Chico, CA	Butte	0.046	0.565	0.011	0.170	0.075	0.061	0.051

*All data up to the 99th percentile of PM2.5 concentration used for RMA analysis.

Table S3.
NOy/CO AERs for each site and NOx/CO ERs calculated from the county emission inventories. The NOy/CO AERs are calculated using an RMA regression of all data at each site. NA means NOy data was not available. The county ERs are calculated for the mobile sources, fuel combustion sources, the sum of all sources, and the sum of all sources except fires. The county ERs are calculated for the sum of all sources, all sources except fires, and all sources except fires and dust.

		NOy/CO AERs (ppbv $ppbv^{-1}$)				NEI11 County NOx/CO ERs (ppbv $ppbv^{-1}$)			
		All data		Weekday rush hour data					
Site location	**Site county**	**slope**	**R^2**	**slope**	**R^2**	**Mobile**	**Fuel combustion**	**Sum of all sources**	**Sum except fires**
Seattle - Beacon Hill	King	0.185	0.711	0.218	0.753	0.180	0.090	0.173	0.173
Seattle - 10th St	King	NA	NA	NA	NA	0.180	0.090	0.173	0.173
Portland, OR	Multnomah	0.088	0.947	0.095	0.876	0.184	0.163	0.178	0.181
Boise, ID	Ada	0.136	0.718	0.158	0.780	0.182	0.188	0.175	0.182
Denver, CO	Denver	0.145	0.801	0.160	0.831	0.166	0.625	0.195	0.195
Stockton, CA	San Joaquin	NA	NA	NA	NA	0.323	0.523	0.344	0.360
Fresno, CA	Fresno	0.070	0.918	0.079	0.812	0.340	0.376	0.306	0.349
Reno, NV	Washoe	0.130	0.858	0.141	0.910	0.169	0.201	0.147	0.157
Chico, CA	Butte	NA	NA	NA	NA	0.286	0.158	0.167	0.256