Table S1. Outer (CONUS) boundary condition concentrations of major aerosol species.

Component	**Concentration** (μg m^{-3})			
	West	East	South	North
Nitrate	0.01	0.01	0.03	0.03
Ammonium	0.14	0.25	0.24	0.16
Sulfate	0.64	1.12	0.81	0.68
Elemental Carbon	0.04	0.05	0.09	0.03
Organic Aerosol (Winter)	0.20	0.16	0.58	0.80
Organic Aerosol (Summer)	0.80	0.80	0.80	0.80

Evaluation of interpolated meteorological data at 1 x 1 km resolution

WRF was evaluated at the METAR stations surrounding the city of Pittsburgh (Figure 1). The analysis focuses on the variables affecting atmospheric chemistry and dispersion. The mean monthly (February, July) diurnal cycle of temperature (T2), relative humidity (RH2) and wind speed (WS10) averaged at the 7 monitoring stations, as observed and as simulated from WRF is presented in Figure 2a.

The cycles are well reproduced for T2 and RH2 in the warm season. This also holds true for the daytime cycle in the cold season; at winter nights however, WRF underestimates (overestimates) T2 (RH2) across all stations (Figure 2b). This results in larger RMSE in February, being 3.1°C for T2 and 18.9% for RH2, i.e., roughly 50% increased with respect to July (Table S1). The simulated wind demonstrates an underestimation tendency during nighttime and an overestimation tendency during daytime, resulting in a mild overestimation in the amplitude of the diurnal cycle. Seasonal errors are comparable (RMSE~1.7m/s).

The spread of errors across stations is larger during (a) nighttime for the thermodynamic variables (nocturnal boundary layer), (b) daytime for the dynamic variables (small-scale winds affected by resolution). Moreover, the phasing is increasing in the order WS10, RH2, T2 and is generally better in February due to the larger impact of the synoptic forcing. No significant differences found spatially.

The above results are consistent with weaker vertical diffusion in the stable boundary layer (night) and stronger vertical momentum fluxes in the convective boundary layer (day). Even such, the magnitude and phasing of the errors are small, making the simulations suitable for air quality studies.

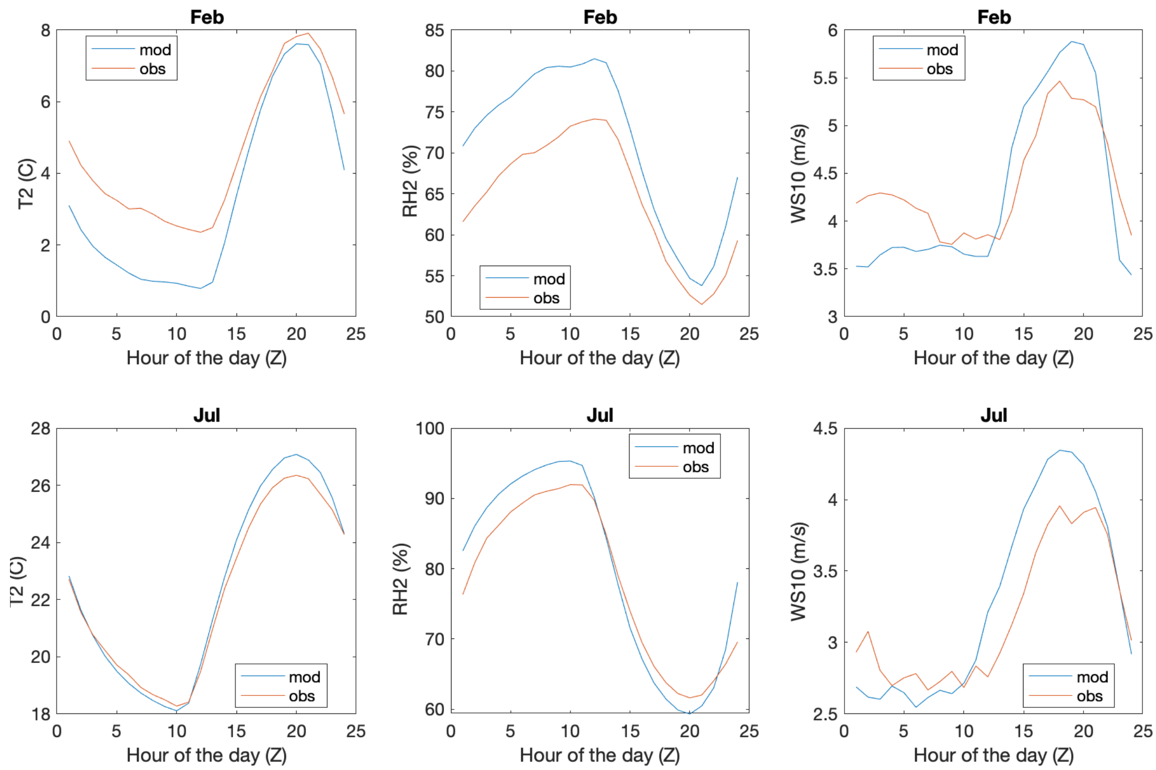

Figure S1. (a) Mean monthly diurnal cycle of temperature (T2), relative humidity (RH2) and wind speed (WS10) averaged at the 7 monitoring stations, as observed and as simulated from WRF. All hours are in UTC.

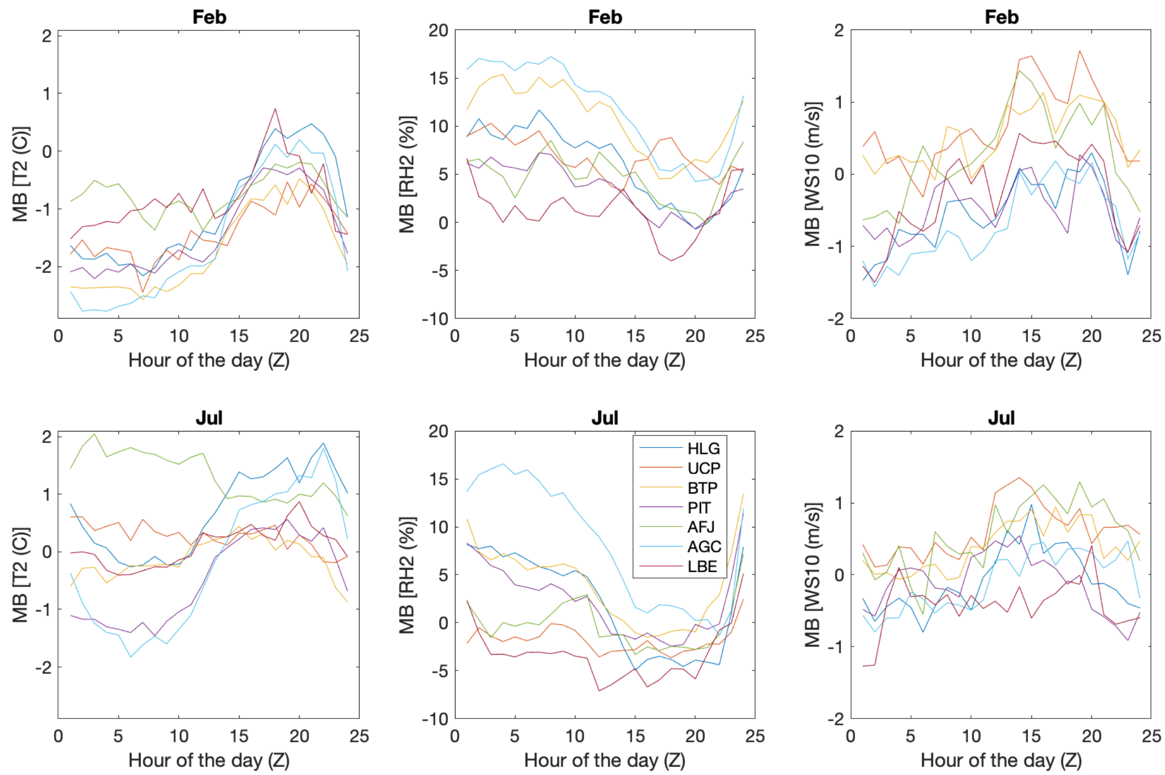

Figure S2. Mean monthly diurnal cycle of the mean bias of temperature, relative humidity, and wind speed at each of the 7 monitoring stations. All hours are in UTC.

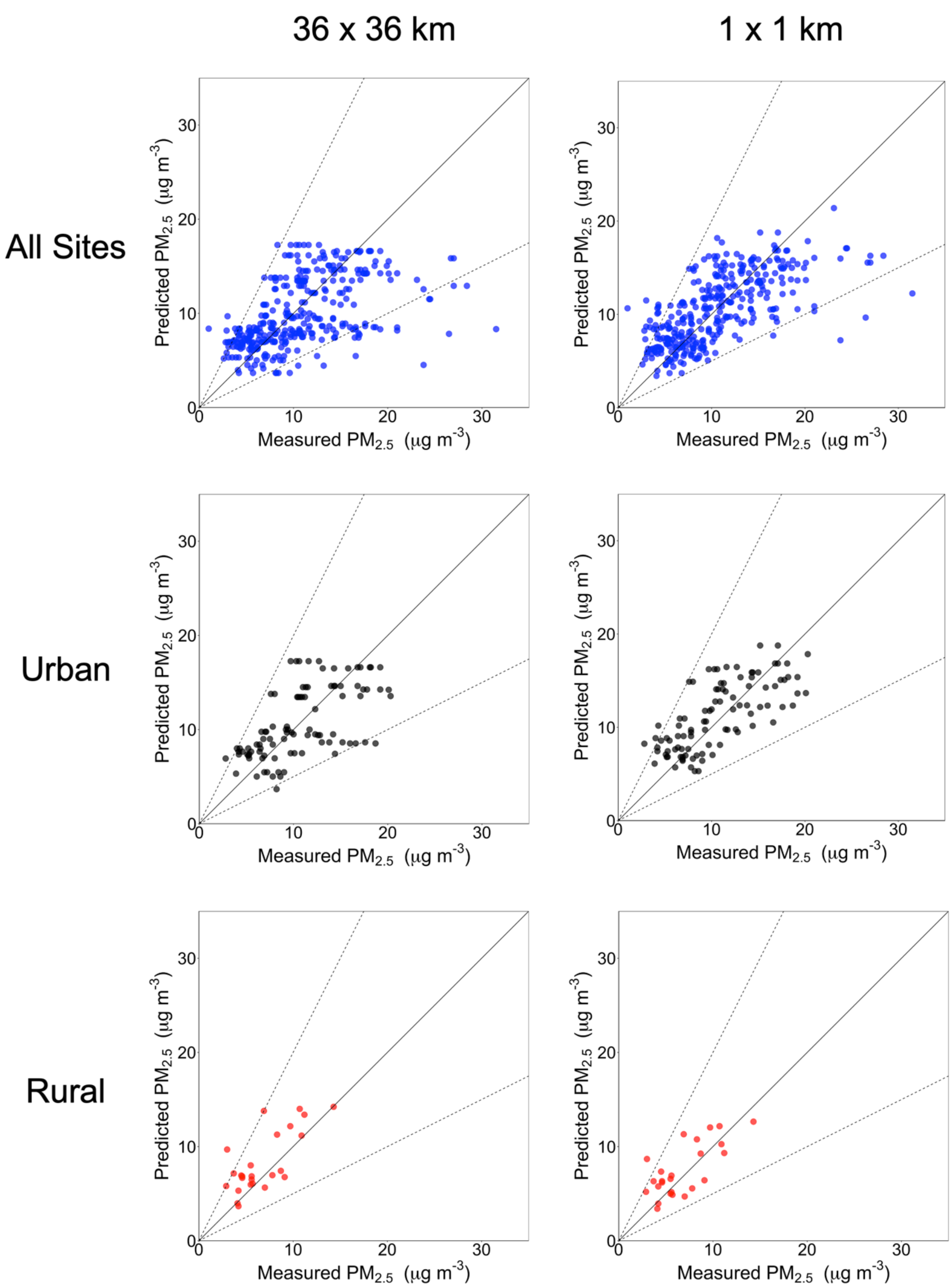

Figure S3. Comparison of PMCAMx-v2.0 predicted concentrations of $PM_{2.5}$ with EPA regulatory measurements in the inner modeling domain at 36 x 36 and 1 x 1 km resolution during February 2017, for all sites, urban sites, and rural sites.

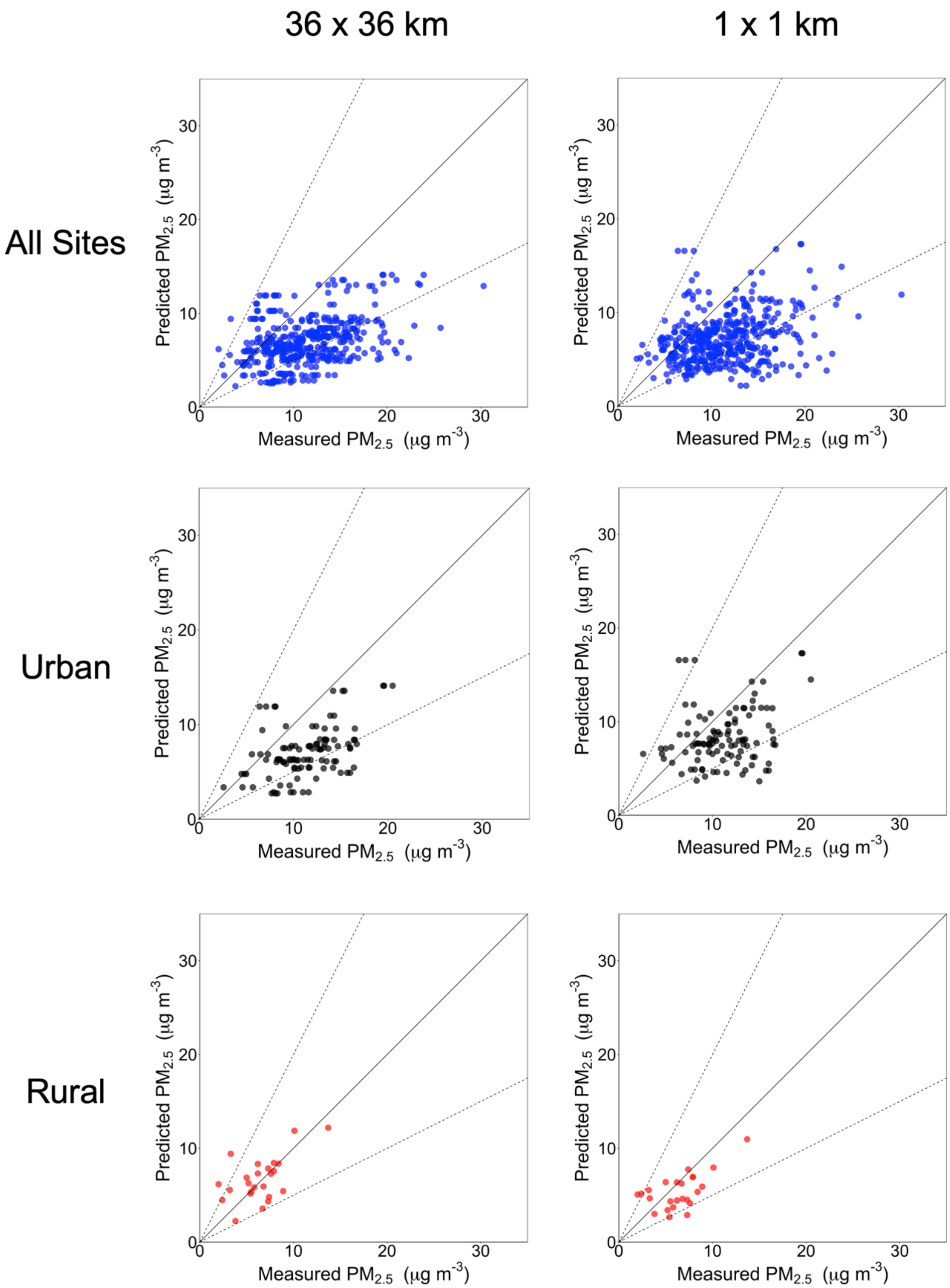

Figure S4. Comparison of PMCAMx-v2.0 predicted concentrations of $PM_{2.5}$ with EPA regulatory measurements in the inner modeling domain at 36 x 36 and 1 x 1 km resolution during July 2017, for all sites, urban sites, and rural sites.

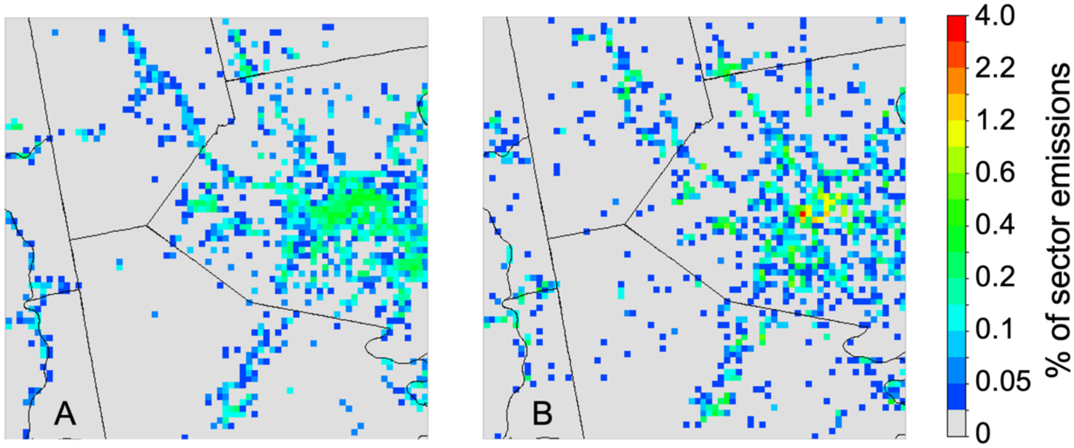

Figure S5. Percentage of sector $PM_{2.5}$ emissions in each 1x1 km computational cell for commercial cooking in February 2017 using: (**A**) old surrogates (**B**) novel surrogates using the normalized restaurant count approach. The values of the colored points in each frame add up to 1.0, corresponding to 100% of emissions for the respective sectors.

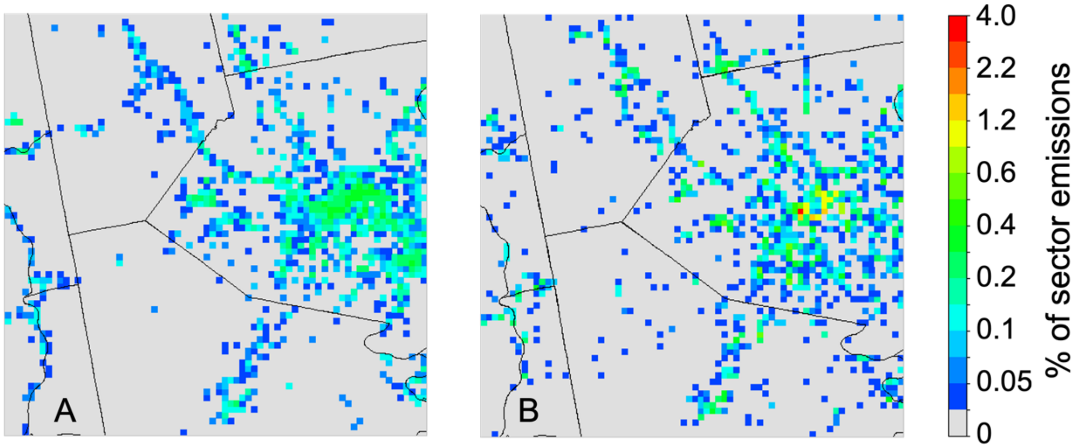

Figure S6. Percentage of sector $PM_{2.5}$ emissions in each 1x1 km computational cell for commercial cooking in July 2017 using: (**A**) old surrogates (**B**) novel surrogates using the normalized restaurant count approach. The values of the colored points in each frame add up to 1.0, corresponding to 100% of emissions for the respective sectors.

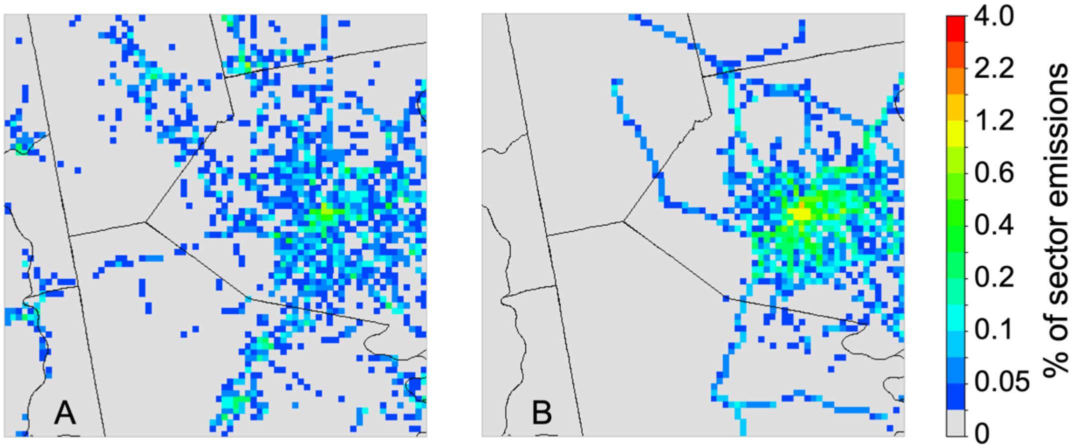

Figure S7. Percentage of sector $PM_{2.5}$ emissions in each 1x1 km computational cell for commercial cooking in February 2017 using: (**A**) old surrogates (**B**) novel surrogates using the simulated traffic approach. The values of the colored points in each frame add up to 1.0, corresponding to 100% of emissions for the respective sectors.

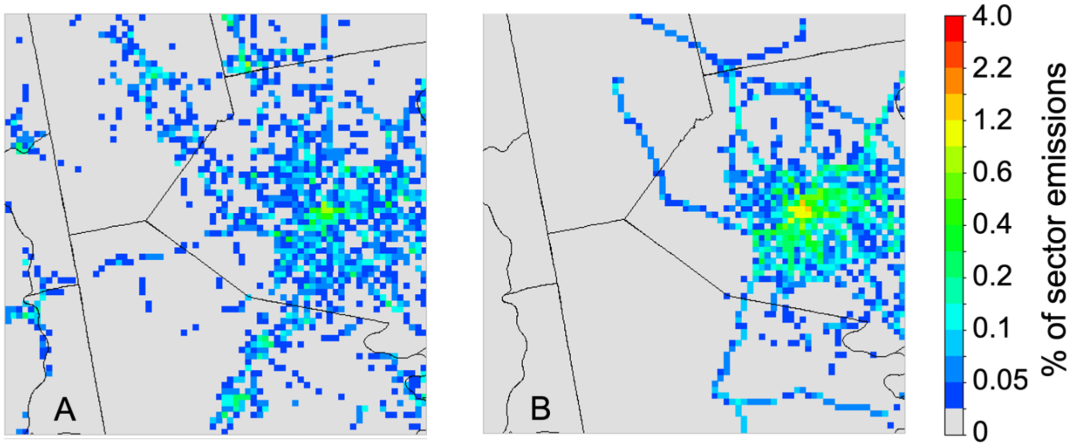

Figure S8. Percentage of sector $PM_{2.5}$ emissions in each 1x1 km computational cell for commercial cooking in February 2017 using: (**A**) old surrogates (**B**) novel surrogates using the simulated traffic approach. The values of the colored points in each frame add up to 1.0, corresponding to 100% of emissions for the respective sectors.