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Dynamics-based estimates of decline trend with fine temporal variations in China's $PM_{2.5}$ emissions

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Supplementary Information

Effects of meteorology

An observing system simulation experiment (OSSE) is performed to investigate the effects of time-varying boundary layer. A nature run is first conducted from 0000 UTC 25 December 2015 to 0000 UTC 2 February 2016, forced by the time-invariant source emissions PR2010 (the true emission). Synthetic observations of the six conventional air pollutant concentrations (i.e., PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3, and CO) are generated from the natural run. Hourly synthetic observations are created from 0000 UTC 29 December 2015 to 0006 UTC 1 February 2016, by interpolating the gridded true surface concentrations to the chemical observation locations with additive random errors of $N(0, R)$. R is the observation error variance, which is calculated by the formula in Elbern et al. (2007). Outputs from the first four days of the natural run are excluded to avoid the transient effect. Then the prior emissions are generated by $\mathbf{F}^{pr} = (1.8 + \delta(x, y, z, t))\mathbf{F}^{tr}$, where $\mathbf{F}^{tr}$ is the true emission, δ is a random number sampled from the normal distribution $N(0,1)$ (Peng et al. 2015). Ensemble data assimilation experiments are conducted from 0000 UTC 29 December to 0006 UTC 1 February 2016. Outputs from the first two days of the OSSE are excluded due to the spin-up.

The magnitude of posterior $PM_{2.5}$ emission is closer to the true emission than the prior. Figure S1 presents the monthly mean diurnal variations of $PM_{2.5}$ emission fraction from the OSSE. It shows that a little larger estimated $PM_{2.5}$ emission fractions occurred in the morning and smaller estimated $PM_{2.5}$ emission fractions occurred in the afternoon, comparing to the time-invariant true emission. But the diurnal variations of $PM_{2.5}$ emission fractions caused by the boundary layer are not as strong as that caused by the emission itself (Figure 7). The reason may be that we have hourly assimilated observations to simultaneously update the chemical concentrations and source emissions. Therefore, the impacts of time-varying boundary layer on the posterior $PM_{2.5}$ emissions are limited.

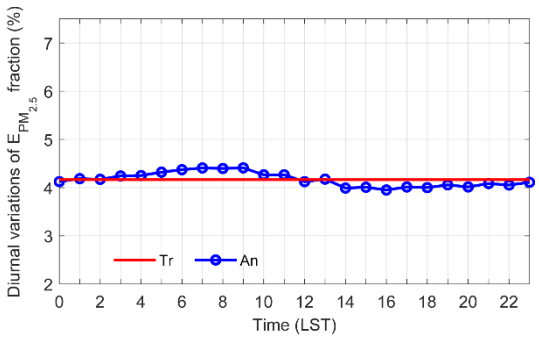

Figure S1. Diurnal variations of $PM_{2.5}$ emission fraction for the Observing System Simulation Experiment.