Supplementary Information

Improving NO_X emissions in Beijing using network observations and a novel perturbed emissions ensemble

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S1. Initial PEE simulations

The initial PEE was constructed using uncertainty ranges determined from the expert elicitation (see Table 1, column *Initial PEE*). The simulations were forced with the same meteorological data and background concentrations of NO and TVOC as the optimised PEE simulations (see Section 2.3). For background NO_2 and O_3, we used baseline concentrations from the long-term monitoring network, defined as the 10^{th} percentile of all values measured in a moving 3-hour window. The difficulties in defining a spatially uniform regional background NO_2 and O_3 are discussed in Section 2.3 and the model sensitivity to different definitions are examined in Section 4.

Figure S1 shows the normalised mean bias factor (NMBF) in NO_2 concentrations output by the initial PEE simulations. The NMBF is an intuitive yet robust measure of the mean magnitude of the factor by which the model outputs differ from the observations and its sense (Yu et al., 2006). It is defined as:

$$NMBF = \frac{\overline{Mod}}{\overline{Obs}} - 1, \text{ if } \overline{Mod} \geq \overline{Obs}$$

$$= 1 - \frac{\overline{Obs}}{\overline{Mod}}, \text{ if } \overline{Mod} < \overline{Obs}$$

where $\overline{Mod}$ and $\overline{Obs}$ represent the means of the modelled and observed values, respectively. As can be seen from Fig. S1, there is a widespread overestimation of annual mean NO_2 concentrations across the long-term monitoring network, spanning different site types. In some simulations, the overestimation is above a factor of 2 (calculated as NMBF+1, when the NMBF is positive) at a few urban sites. More importantly, the entire ensemble of simulations overestimates the annual mean NO_2 concentrations at 19 sites. This is an indication that NO_X emissions are generally high biased in the initial PEE.

The modelled annual mean diurnal variations of NO_2 concentrations are often much higher than those observed (Fig. S2), providing further indications of a high bias in the NO_X emissions. In addition, the modelled diurnal profiles at most sites are characterised by two peaks that coincide with the peak traffic periods, while such peaks are less distinct, if not completely absent in the observed profiles. This suggests specifically that the transport sector NO_X emissions are overestimated.

In summary, the initial PEE simulations failed to produce sufficient modelled concentrations comparable to the observations, such that the elicited uncertainty ranges in NO_X emissions could not be reduced. Hence, we constructed an optimised PEE described in Section 2.2.

Table S1. Long-term air quality monitoring sites in operation in 2016 and located within the modelling domain.

Name	Acronym	Longitude (°E)	Latitude (°N)	Type
Dongsi	DS	116.417	39.929	Urban site
Tiantan	TT	116.407	39.886	Urban site
Guanyuan	GY	116.339	39.929	Urban site
Wanshouxigong	WSXG	116.352	39.878	Urban site
Aotizhongxin	ATZX	116.397	39.982	Urban site
Nongzhanguan	NZG	116.461	39.937	Urban site
Wanliu	WL	116.287	39.987	Urban site
Beibuxinqu	BBXQ	116.174	40.09	Urban site
Zhiwuyuan	ZWY	116.207	40.002	Urban site
Fengtaihuayuan	FTHY	116.279	39.863	Urban site
Yungang	YG	116.146	39.824	Urban site
Gucheng	GC	116.184	39.914	Urban site
Qianmen	QM	116.395	39.899	Traffic monitoring site
Yongdingmennei	YDMN	116.394	39.876	Traffic monitoring site
Xizhimenbei	XZMB	116.349	39.954	Traffic monitoring site
Nansanhuan	NSH	116.368	39.856	Traffic monitoring site
Dongsihuan	DSH	116.483	39.939	Traffic monitoring site
Fangshan	FS	116.136	39.742	Suburban site
Daxing	DX	116.404	39.718	Suburban site
Yizhuang	YZ	116.506	39.795	Suburban site
Tongzhou	TZ	116.663	39.886	Suburban site
Shunyi	SY	116.655	40.127	Suburban site
Changping	CP	116.23	40.217	Suburban site
Mentougou	MTG	116.106	39.937	Suburban site
Pinggu	PG	117.1	40.143	Suburban site
Huairou	HR	116.628	40.328	Suburban site
Miyun	MY	116.832	40.37	Suburban site
Dingling	DL	116.22	40.292	Clean site
Badaling	BDL	115.988	40.365	Regional background site
Donggaocun	DGC	117.12	40.1	Regional background site
Yongledian	YLD	116.783	39.712	Regional background site
Yufa	YF	116.3	39.52	Regional background site
Liulihe	LLH	116	39.58	Regional background site

Table S2. Low-cost SNAQ (Sensor Network for Air Quality) deployed for near-surface measurement within the modelling domain during the APHH-Beijing winter campaign (November-December 2016).

ID	Longitude (°E)	Latitude (°N)
SNAQ03	117.393	40.164
SNAQ04	117.423	40.164
SNAQ07	117.419	40.163
SNAQ10	116.367	39.970
SNAQ12	117.405	40.166
SNAQ15	116.402	39.750
SNAQ16	116.425	39.942
SNAQ17	116.395	39.981
SNAQ21	116.391	40.272
SNAQ22	116.360	39.830
SNAQ23	116.557	39.746
SNAQ25	116.300	39.968
SNAQ26	116.661	40.360
SNAQ28	116.318	39.621
SNAQ29	116.363	39.978
SNAQ31	116.492	39.997
SNAQ34	116.233	39.642
SNAQ35	116.414	40.128
SNAQ36	116.680	40.396
SNAQ37	116.428	40.158
SNAQ38	116.273	39.725
SNAQ39	116.371	39.974

Table S3. Different definitions of background concentrations of NO_2 and O_3 used in the background sensitivity simulations. Upwind concentration refers to the inverse distance weighted mean concentration of the two clean or regional background sites in the upwind direction of each hour. 10^{th} and 90^{th} percentile concentrations refer to the 10^{th} and 90^{th} concentration of all sites in a moving 3-h window. In the optimised PEE simulations, upwind concentration is used for both background NO_2 and O_3.

Background sensitivity simulations	Background NO_2	Background O_3
S1	Upwind concentration	90^{th} percentile concentration
S2	Upwind concentration	10^{th} percentile concentration
S3	10^{th} percentile concentration	Upwind concentration
S4	10^{th} percentile concentration	90^{th} percentile concentration
S5	10^{th} percentile concentration	10^{th} percentile concentration

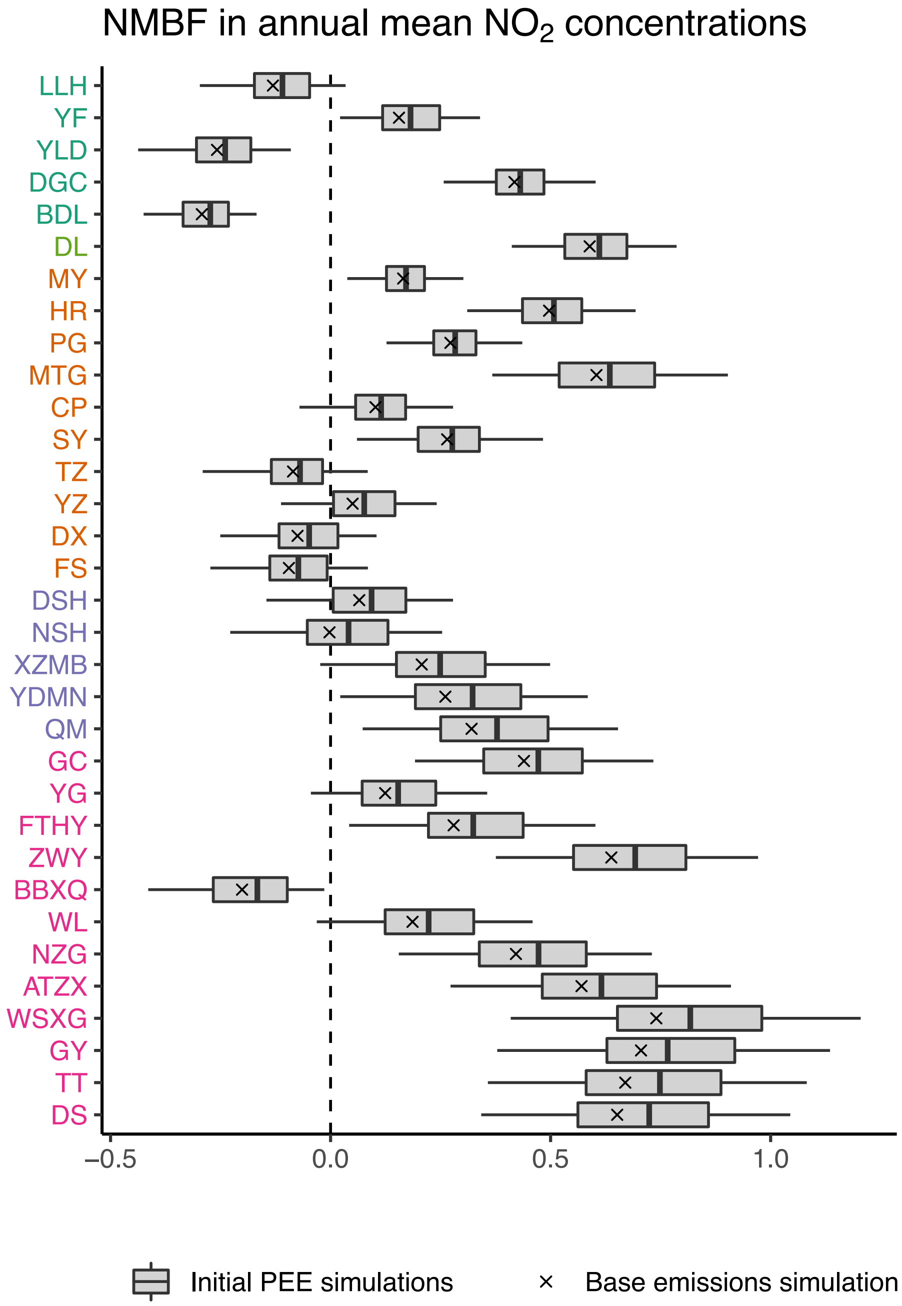

Figure S1. Distribution of the normalised mean bias factors (NMBF) in annual mean NO_2 concentrations associated with the initial perturbed emissions ensemble (PEE) simulations and the simulation forced with the base emissions at each long-term monitoring site. The monitoring sites are colour-coded according to the site type: urban site (magenta), traffic monitoring site (purple), suburban site (orange), clean site (light green) and regional background site (green).

Figure S2. Annual mean diurnal variations of NO_2 concentrations simulated with the initial perturbed emissions ensemble (PEE) and with the base emissions, compared to the observations at each long-term monitoring site. The monitoring sites are colour-coded according to the site type: urban site (magenta), traffic monitoring site (purple), suburban site (orange), clean site (light green) and regional background site (green). Note the different scales on the y-axis used in the panels.

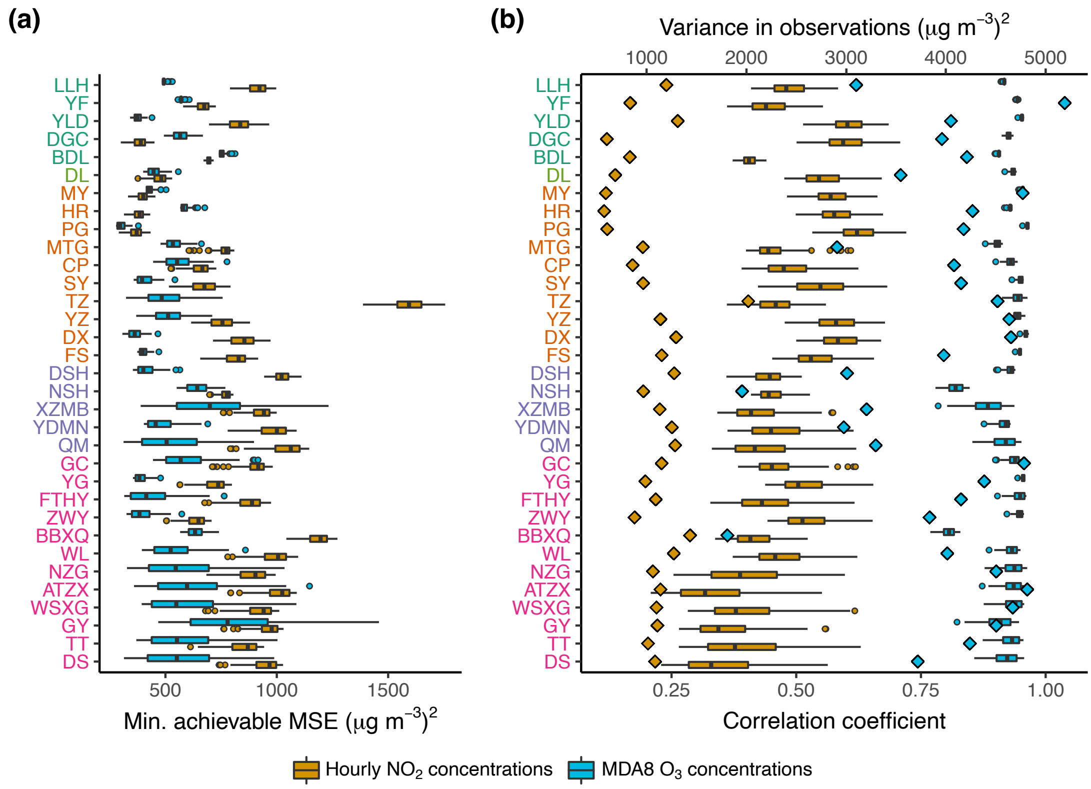

Figure S3. Distribution of (a) the minimum achievable mean square errors (MSE) and (b) the Pearson's correlation coefficient in hourly NO_2 concentrations and daily maximum 8-hour mean (MDA8) O_3 concentrations associated with the optimised perturbed emissions ensemble simulations a at each long-term monitoring site. The observed variance in hourly NO_2 concentrations and MDA8 O_3 concentrations are also shown in panel (b) by diamond symbols against the secondary x-axis (at the top). The monitoring sites are colour-coded according to the site type: urban site (magenta), traffic monitoring site (purple), suburban site (orange), clean site (light green) and regional background site (green). Note the different scales on the y-axis used in the panels.

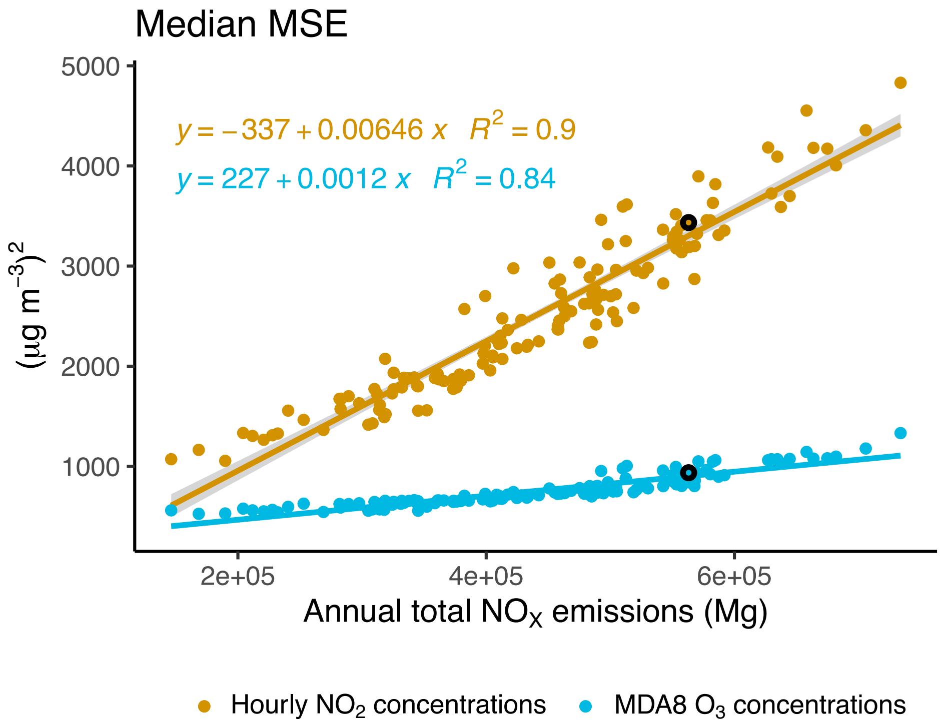

Figure S4. Median mean square errors (MSE) in hourly NO_2 concentrations and daily maximum 8-hour mean (MDA8) O_3 concentrations associated with the optimised perturbed emissions ensemble (PEE) simulations and the base run (marked with black strokes), as a function of the input annual total NO_X emissions. The fitted linear regression models and the coefficients of determination (R^2) are shown in the corresponding colour. The confidence intervals are shown by grey shading.

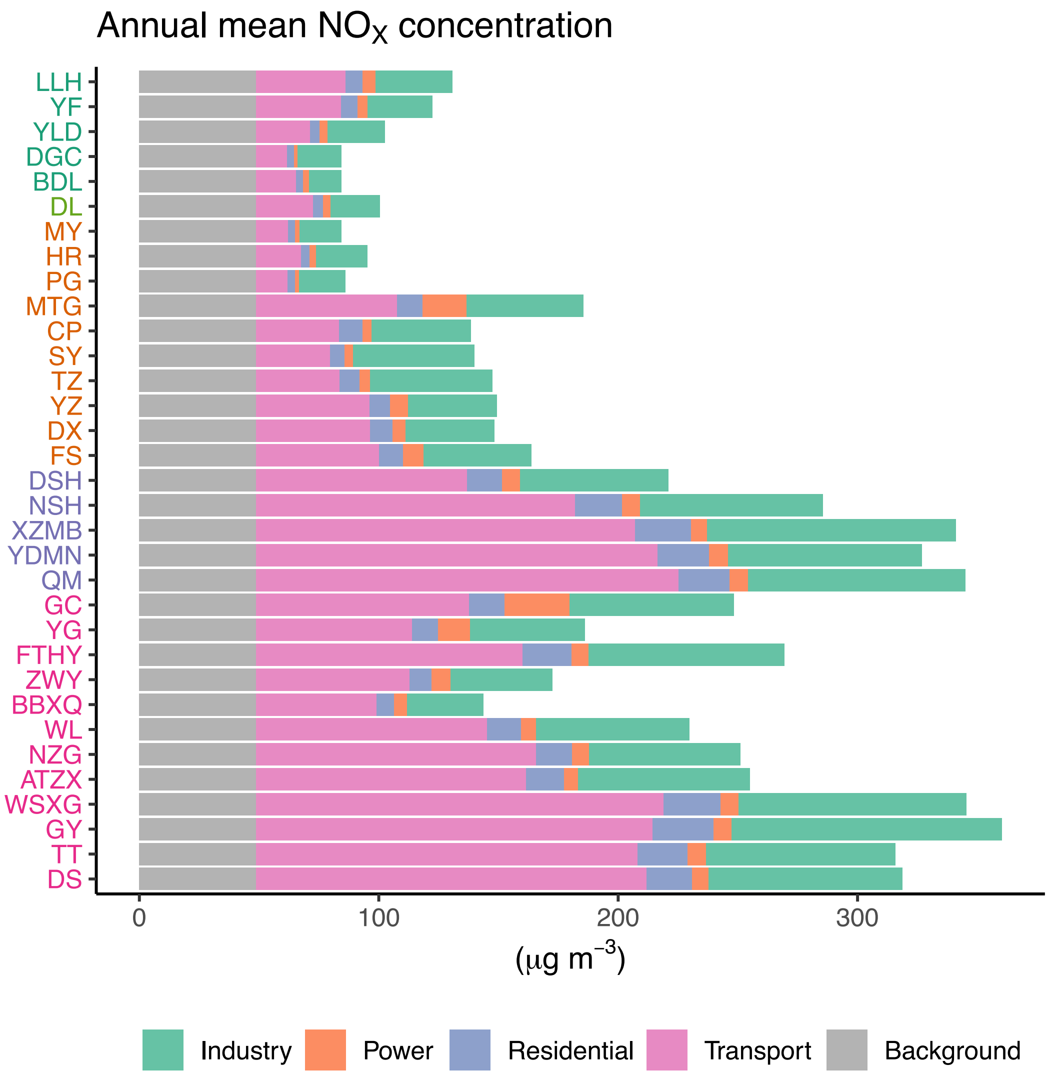

Figure S5. Annual mean NO_X concentrations at each long-term monitoring site simulated with the base emissions, shown as contributions of NO_X emissions from individual source sectors and the input background concentration. Chemistry calculations were disabled in the source apportionment, thus it is more meaningful to examine concentrations of NO_X than those of NO_2. NO_X concentrations (in mass units) are calculated by assuming that 100% of the NO_X is NO_2. The monitoring sites are colour-coded according to the site type: urban site (magenta), traffic monitoring site (purple), suburban site (orange), clean site (light green) and regional background site (green).

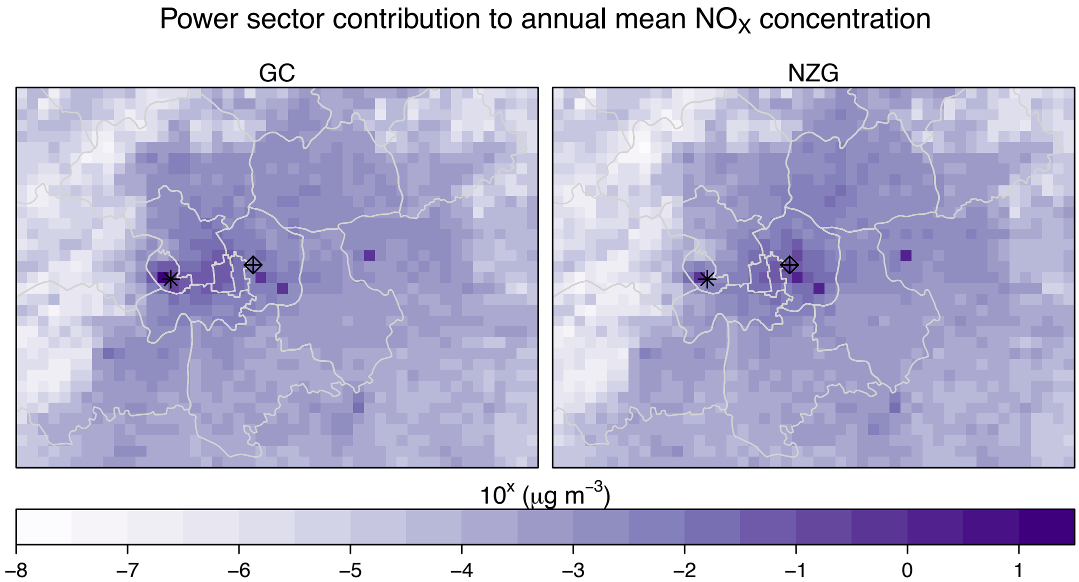

Figure S6. Contribution of the power sector NO_X emissions from each grid cell to the annual mean NO_X concentrations at the sites GC (circled plus symbol) and NZG (diamond plus symbol) in the base run. The contributions are log transformed (base 10) due to a strong positive skewness (i.e. the colour scale shows the exponent). NO_X concentrations (in mass units) are calculated by assuming that 100% of the NO_X is NO_2.

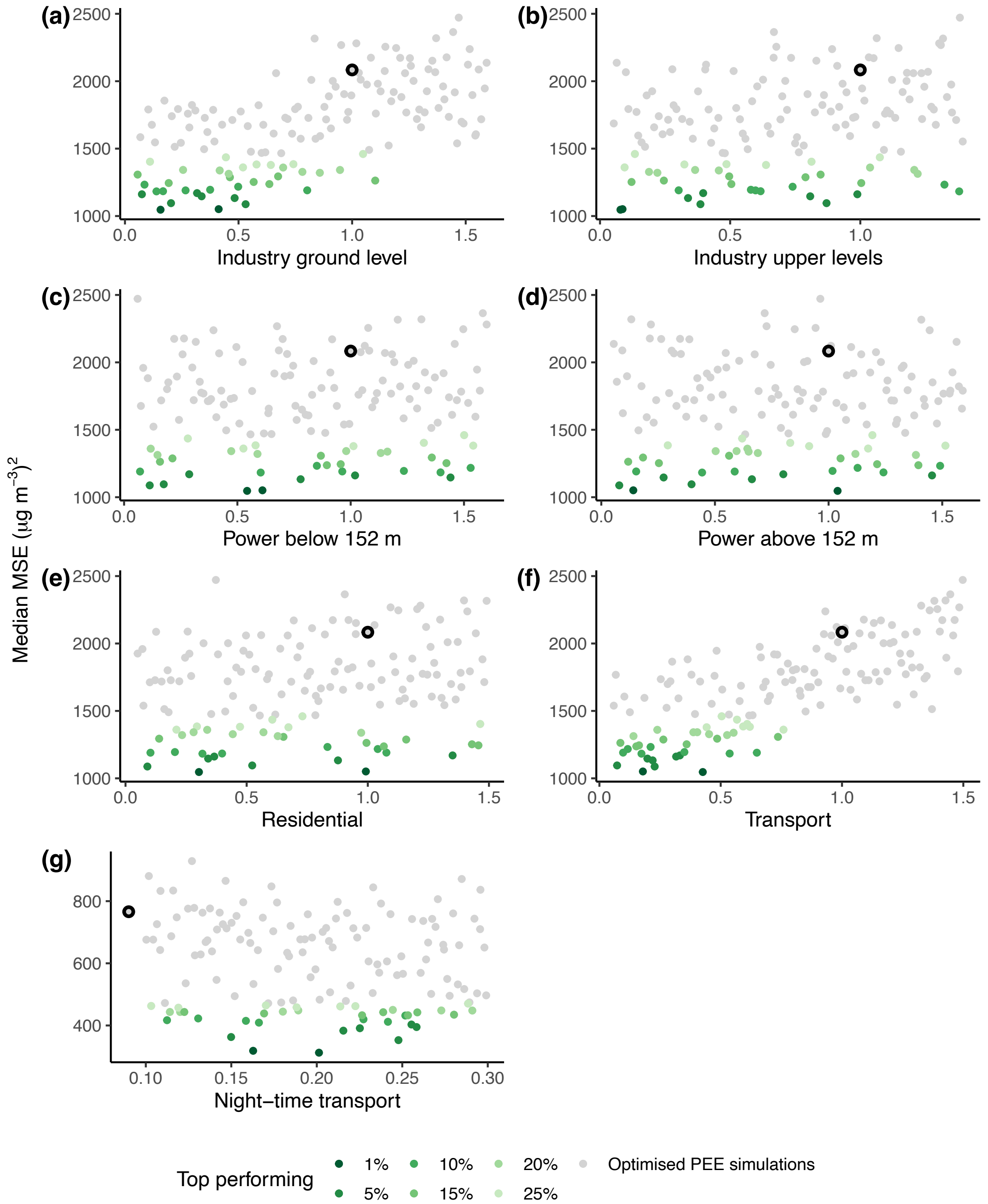

Figure S7. Average performance of the optimised perturbed emissions ensemble (PEE) simulations and the base run (marked with black strokes) as a function of emission parameter values. The scales on the x-axes correspond to the uncertainty ranges in Table 1. The top performing 25%, 20%, 15%, 10%, 5% and 1% of the simulations are coloured in a darkening green shade, as measured by their median mean square errors (MSE) in hourly NO_2 concentrations at the SNAQ sites across the modelling domain in all panels except in (g), where median mean square errors in the mean diurnal variations of NO_2 concentrations (during the campaign period) are used (note the different scale on the y-axis).

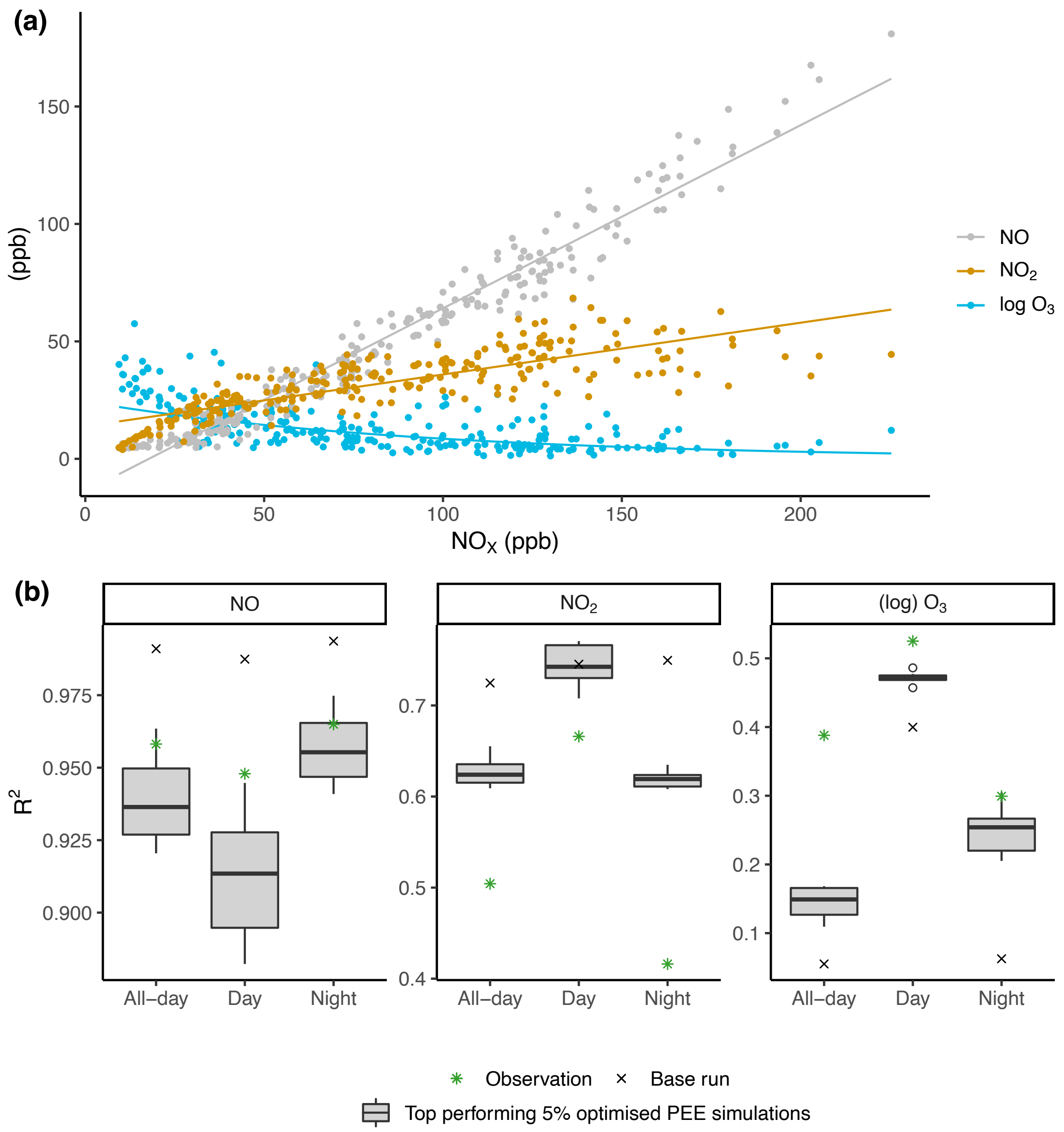

Figure S8. (a) Observed mixing ratios of NO, NO_2 and log transformed O_3 as a simple linear function of NO_X mixing ratios at all SNAQ sites. Daily mean mixing ratios are shown for clarity, but the models are fitted to hourly data. (b) Distributions of the coefficients of determination (R^2) of the linear regression models fitted between hourly mixing ratios of NO, NO_2, log transformed O3 and those of NO_X output at all SNAQ sites by the top performing 5% of the optimised perturbed emissions ensemble (PEE) simulations and the base run, compared to the corresponding R^2 values of the models fitted to the SNAQ measurements. Daytime is defined as complete hours between sunrise and sunset in Beijing during November-December 2016, namely 8:00-15:00 local time.