Supplementary Information

Improving NOX emission estimates in Beijing using network observations and a 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 adjusted PEE simulations (see Section 2.3). For background NO2 and O3, we used baseline concentrations from the long-term monitoring network, defined as the 10th percentile of all values measured in a moving 3-hour window. The difficulties in defining a spatially uniform regional background NO2 and O3 are discussed in Section 2.3 and the model sensitivity to different definitions are examined in Section 4.

Figure S2 shows the normalised mean bias factor (NMBF) in NO2 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:

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

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

where $ \overbar{Mod}$ and $\overbar{Obs}$ represent the means of the modelled and observed values, respectively. As can be seen from Fig. S2, there is a widespread overestimation of annual mean NO2 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 NO2 concentrations at 19 sites. This is an indication that NOX emissions are generally high biased in the initial PEE.

The modelled annual mean diurnal variations of NO2 concentrations are often much higher than those observed (Fig. S2), providing further indications of a high bias in the NOX 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 NOX 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 NOX emissions could not be reduced. Hence, we constructed an adjusted 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 NO2 and O3 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. 10th and 90th percentile concentrations refer to the 10th and 90th concentration of all sites in a moving 3-h window. In the adjusted PEE simulations, upwind concentration is used for both background NO2 and O3.

|  |  |  |
| --- | --- | --- |
| Background sensitivity simulations | Background NO2 | Background O3 |
| S1 | Upwind concentration  | 90th percentile concentration  |
| S2 | Upwind concentration  | 10th percentile concentration |
| S3 | 10th percentile concentration  | Upwind concentration  |
| S4 | 10th percentile concentration  | 90th percentile concentration  |
| S5 | 10th percentile concentration | 10th percentile concentration |



**Figure S1.** Annual NOX emissions from each source sector and grid cell (of 3 km × 3 km resolution) in the base emissions. For the industry and power sectors, emissions from all vertical layers are aggregated. The administrative divisions of Beijing are shown by light grey outlines.

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**Figure S2.** Distribution of the normalised mean bias factors (NMBF) in annual mean NO2 concentrations associated with (a) the initial and (b) the adjusted perturbed emissions ensemble (PEE) simulations at each long-term monitoring site. In each panel, the simulation forced with the base emissions is also shown. Note that as different background levels of NO2 and O3 are input (in accordance with the initial and the adjusted PEE simulations), these two simulations are not identical, also indicated by the different NMBFs. 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). The circle at the leftmost end of the boxplot for YLD in panel (b) represents an outlying PEE simulation (i.e. with a NMBF outside 1.5 times the interquartile range below the lower quartile).



**Figure S3.** Annual mean diurnal variations of NO2 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.

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**Figure S4.** Distribution of (a) the minimum achievable mean square errors (MSE) and (b) the Pearson’s correlation coefficient in hourly NO2 concentrations and daily maximum 8-hour mean (MDA8) O3 concentrations associated with the adjusted perturbed emissions ensemble simulations a at each long-term monitoring site. The observed variance in hourly NO2 concentrations and MDA8 O3 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 S5.** Median mean square errors (MSE) in hourly NO2 concentrations and daily maximum 8-hour mean (MDA8) O3 concentrations associated with the adjusted perturbed emissions ensemble (PEE) simulations and the base run (marked with black strokes), as a function of the input annual total NOX emissions. The fitted linear regression models and the coefficients of determination (R2) are shown in the corresponding colour. The confidence intervals are shown by grey shading.



**Figure S6.** Annual mean NOX concentrations at each long-term monitoring site simulated with the base emissions, shown as contributions of NOX 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 NOX than those of NO2. NOX concentrations (in mass units) are calculated by assuming that 100% of the NOX is NO2. 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 S7.** Contribution of the power sector NOX emissions from each grid cell to the annual mean NOX 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). NOX concentrations (in mass units) are calculated by assuming that 100% of the NOX is NO2.



**Figure S8.** Average performance of the adjusted 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 NO2 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 NO2 concentrations (during the campaign period) are used (note the different scale on the y-axis).

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**Figure S9.** (a) Observed mixing ratios of NO, NO2 and log transformed O3 as a simple linear function of NOX 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 (R2) of the linear regression models fitted between hourly mixing ratios of NO, NO2, log transformed O3 and those of NOX output at all SNAQ sites by the top performing 5% of the adjusted perturbed emissions ensemble (PEE) simulations and the base run, compared to the corresponding R2 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.