S1 Changes to the model code

The WRF-Chem code file dry_dep_driver.F (v.3.8.1) was changed in order to also allow for increased nighttime mixing in grid cells with high emissions in case an urban physics scheme is used (starting from line 685):

```
     if (p_e_co >= param_first_scalar )then
! if (sf_urban_physics .eq. 0 ) then
         if (emis_ant(i,kts,j,p_e_co) .gt. 0) then
          ekmfull(kts:kts+10) = max(ekmfull(kts:kts+10),1.)
         endif
         if (emis_ant(i,kts,j,p_e_co) .gt. 200) then
          ekmfull(kts:kte/2) = max(ekmfull(kts:kte/2),2.)
         endif
         if (p_e_pm25i > param_first_scalar )then
          if (emis_ant(i,kts,j,p_e_pm25i)+ emis_ant(i,kts,j,p_e_pm25j)
           .GT. 8.19e-4*200) then
           ekmfull(kts:kte/2) = max(ekmfull(kts:kte/2),2.)
          endif
         endif
         if (p_e_pm_25 > param_first_scalar )then
          if (emis_ant(i,kts,j,p_e_pm_25) .GT. 8.19e-4*200) then
           ekmfull(kts:kte/2) = max(ekmfull(kts:kte/2),2.)
          endif
! endif
         endif
```

S2 Emission processing

S2.1 Downscaling

We used TNO-MACC III emission data and in cooperation with TNO downscaled the data from a horizontal resolution of ca. 7kmx7km to a ca. 1kmx1km. We based the downscaling on proxy data, including population density (Environment Database of the Berlin Senate Department for the Environment, Transport and Climate Protection, Landscan 2010 data), traffic density for the area of Berlin (Environment Database of the Berlin Senate Department for the Environment, Transport and Climate Protection) and the road network of Brandenburg (OpenStreetMap). Population data is used to downscale emissions from residential combustion (SNAP2) and product use (SNAP6), traffic data is used to downscale emissions from traffic (SNAP 71-75). The 1kmx1km emission grid is defined so that each coarse grid cell of 7kmx7km is divided into 7x7 parts. From each of the proxy datasets a factor is then calculated indicating the proportion of each proxy data type in one high resolution grid cell within one coarse grid cell. These factors are used in order to downscale the respective emissions in the respective area.

S2.2 Modification of airport emissions for Berlin

Airport emissions in Berlin, designated by point sources within non-road transport emissions in the TNO-MACC III inventory, are split into airport emissions into emissions on the ground and emissions from the LTO-cycle. We attribute 60% of the emissions to emissions on the ground, and the remaining emissions to emissions from the LTO cycle, where we distribute the emissions equally into all layers below 900m. The LTO-cycle includes emissions from takeoff, landing and aircraft cruise up to ca. 900m. Furthermore, the TNO-MACC III inventory still includes emissions from the Berlin-Tempelhof airport, which has been closed to air traffic in 2008. In addition, emissions from Tegel airport seemed unrealistically larger than emissions from Schönefeld airport. Thus we summed the emissions from all three major airports in the Berlin-Brandenburg region included in the TNO-MACC III inventory and re-distributed them onto the two airports active in 2014, based on activity data, attributing 75% of the emissions to Tegel and 25% to Schönefeld.

S3 Model evaluation

S3.1 Statistical indicators

The statistical indicators used in this study include the mean bias (MB), normalized mean bias (NMB), root mean square error (RMSE) and Pearson correlation coefficient (R) and are defined as follows, with the model results M, observations O, number of model-observations pairs N and standard deviation σ:

$$MB = \sum_{i=1}^{N}(M_i - O_i) \tag{1}$$

$$NMB = \frac{\sum_{i=1}^{N}(M_i - O_i)}{\sum_{i=1}^{N} O_i} \tag{2}$$

$$RMSE = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(M_i - O_i)} \tag{3}$$

$$R = \frac{\frac{1}{N}\sum_{i=1}^{N}(M_i - \overline{M})(O_i - \overline{O})}{\sigma_M \sigma_O} \tag{4}$$

S3.2 Additional model performance indicators

In addition to the model quality objective (MQO), performance indicators for the mean bias and normalized mean bias are indicated in the manuscript and defined as follows, following Pernigotti et al. (2013):

$$|NMB| < \frac{2RMS_U}{\overline{O}} \tag{5}$$

$$|MB| < 2U(\overline{O}) \tag{6}$$

With the root mean square of the measurement uncertainty

$$RMS_U = \sqrt{\frac{1}{N}\sum_{i=1}^{N} U_{O_i}^2} \tag{7}$$

and $U(\overline{O})$ the uncertainty of the mean of the observed time series.

S4 Spectral decomposition

Observed and modelled time series are spectrally decomposed into a long term (LT), synoptic (SY), diurnal (DU) and intra-diurnal (ID) component, following Hogrefe et al. (2000), and Galmarini et al. (2013). A Kolmogorov-Zurbenko filter $kz_{m,k}$ was used with the time windows m and smoothing parameter k, time series x and time t:

$$
\begin{aligned}
ID(t) &= x(t) - kz_{3,3}(x(t)) && (8)\\
DU(t) &= kz_{3,3}(x(t)) - kz_{13,5}(x(t)) && (9)\\
SY(t) &= kz_{13,5}(x(t)) - kz_{103,5}(x(t)) && (10)\\
LT(t) &= kz_{103,5}(x(t)) && (11)
\end{aligned}
$$

The decomposition was done in R, using the library *kza*.

S5 Supplementary figures

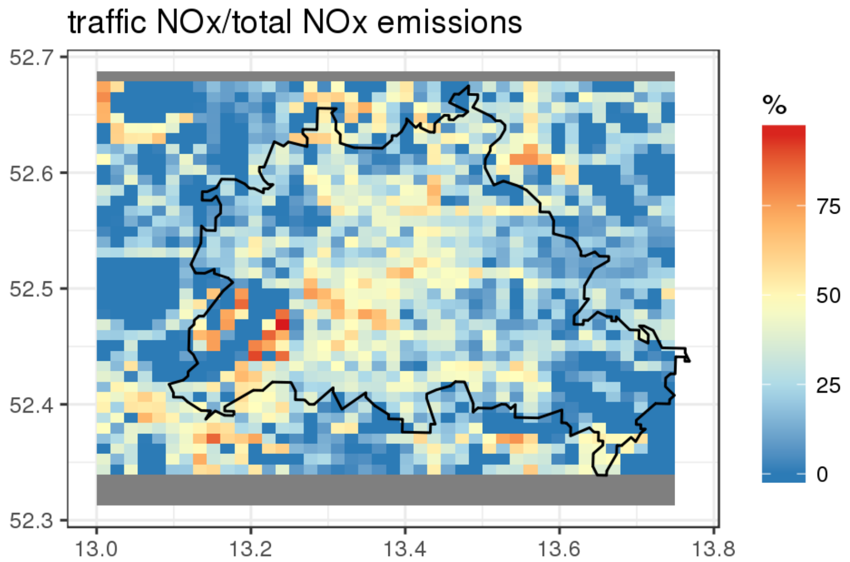

Figure S1. Contribution of traffic NOx emissions to total annual surface NOx emissions in the Berlin-Brandenburg area, based on the downscaled version of TNO-MACC III.

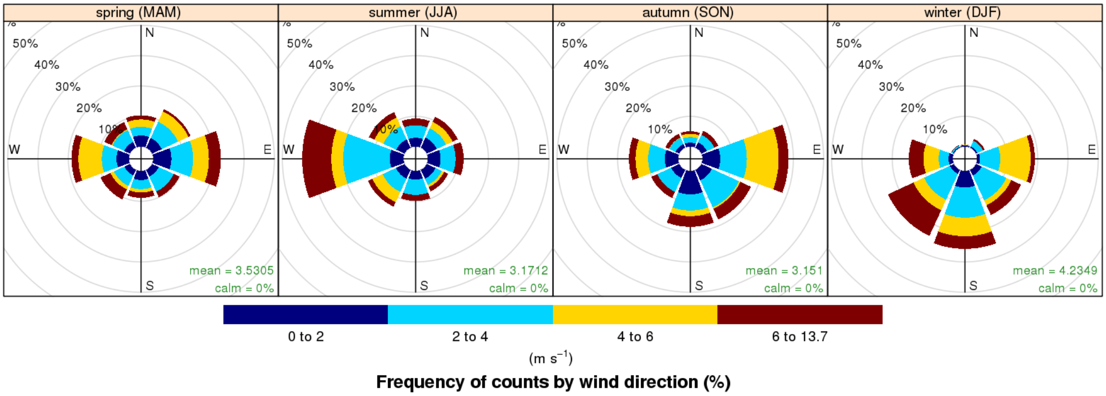

Figure S2. Wind rose showing the frequency distribution of wind speed and direction for the Berlin DWD stations, observations.

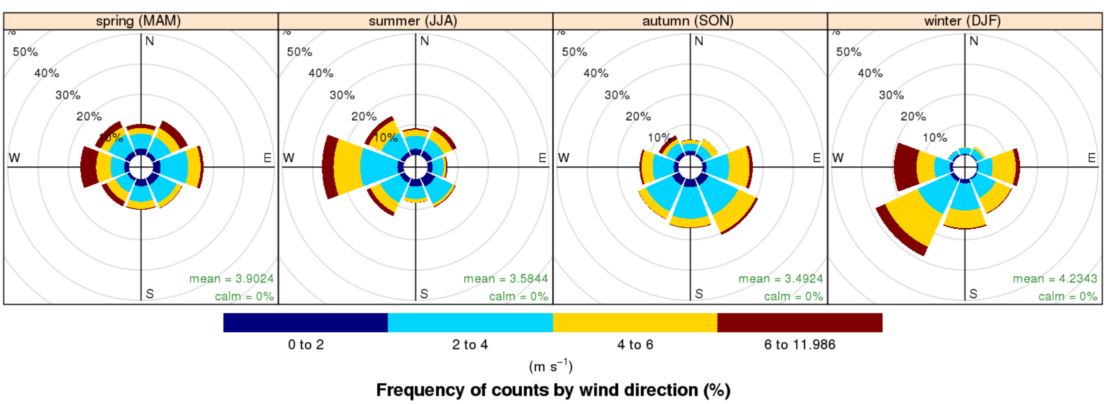

Figure S3. Wind rose showing the frequency distribution of wind speed and direction for the Berlin DWD stations, model results.

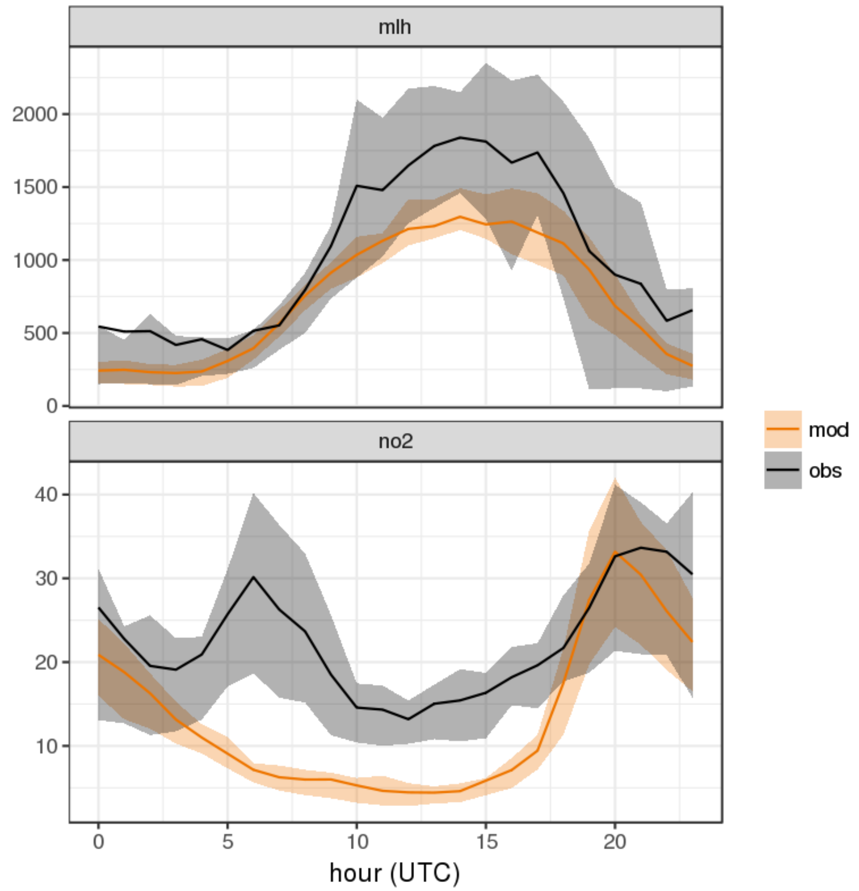

Figure S4. Mean diurnal cycles of observed (obs) and modelled (mod) mixing layer height (MLH) and NO_2 concentrations at Nansenstraße. All data are only averaged over times when MLH observed with a ceilometer is available. This includes between 24-57 hourly values between 20 June and 27 August 2014. The shaded areas show the 25th and 75th percentiles of the data. MLH is given in m, NO_2 concentrations are given in μg m^{-3}.

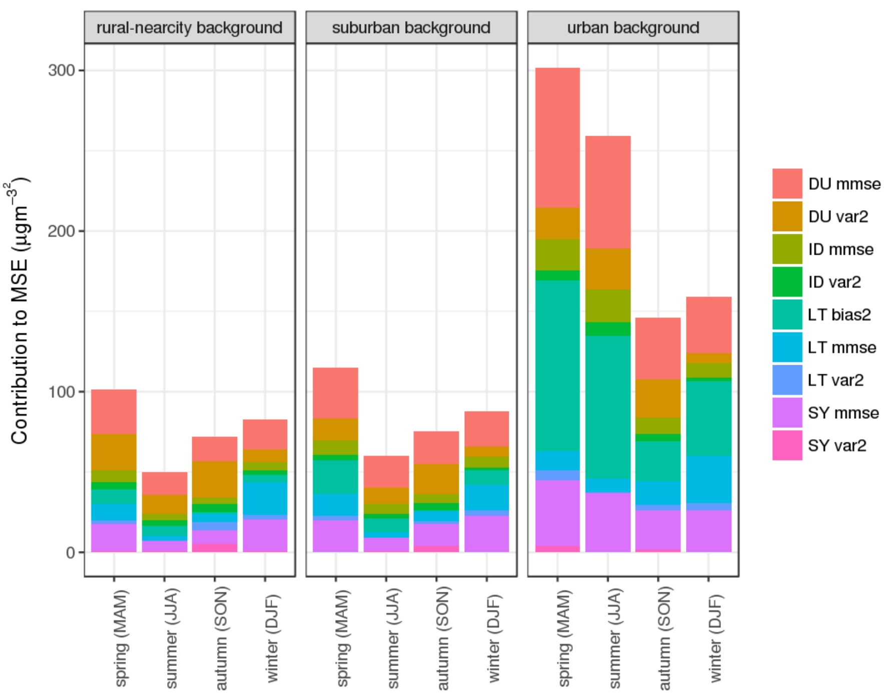

Figure S5. Contribution to mean square error of model results per season and station class.

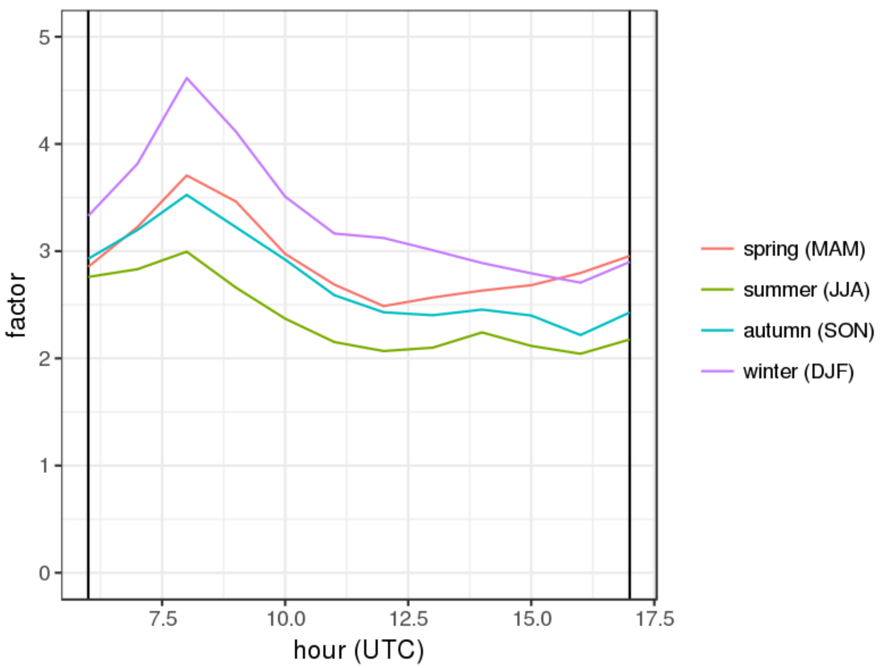

Figure S6. Time- and season-dependent NO_x-emission correction factor.