Supplementary materials

Evaluating emissions and meteorological contributions to air quality trends in northern China based on measurements at a regional background station

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Random Forest (RF) model parameters:

• A forest of 200 tress, n_tree=200;

• Number of times to sample data and then predict, n_samples=300;

• The number of variables that may split at each node, Mtry=4;

• The minimum size of nodes size, min_node_size=3

Model performance's evaluation

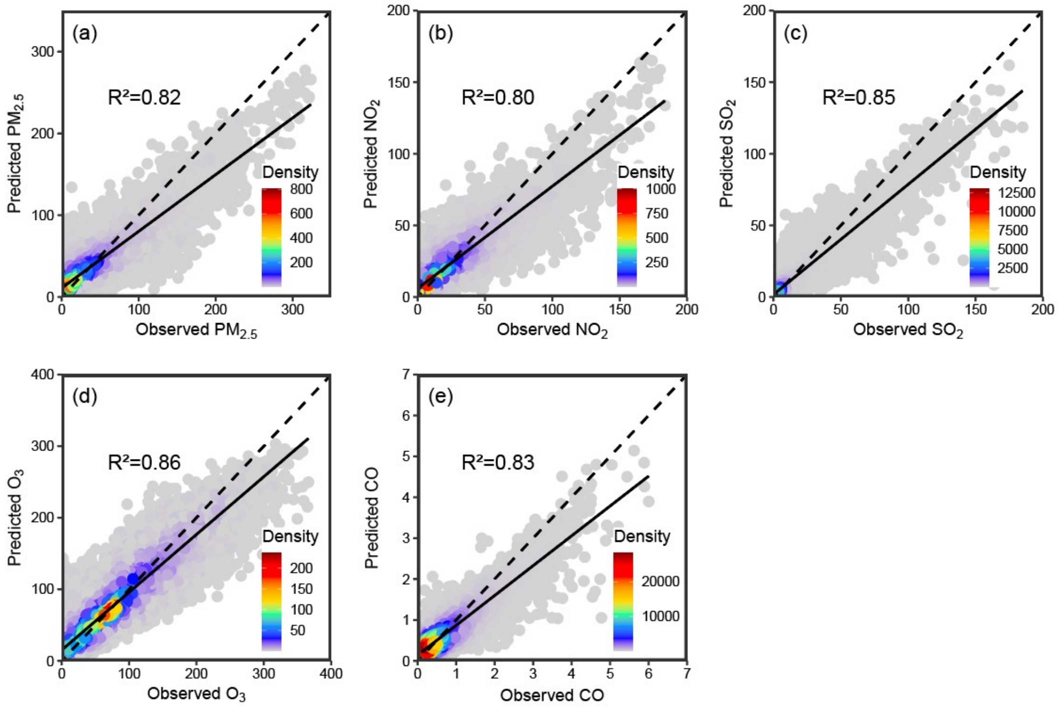


Fig.S1. Correlations between hourly observed and predicted data from training data sets (the units of $PM_{2.5}$, NO_2, SO_2, and O_3 are μg m^{-3} while CO is mg m^{-3}).

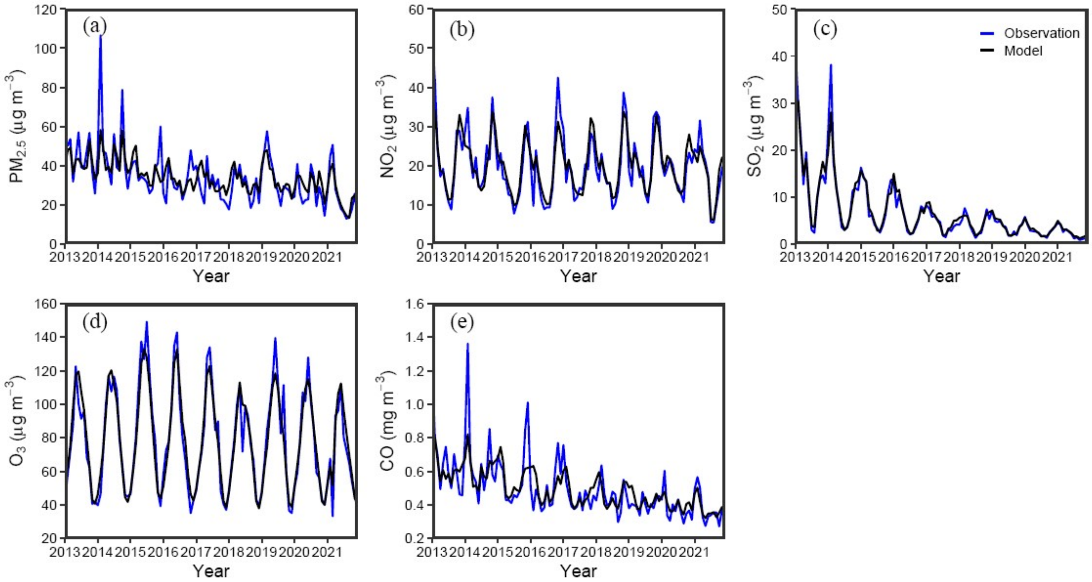


Fig. S2. The variations of monthly averaged pollutants before and after normalization (meteorology-adjusted). "Model" in the figure means the modelled concentration of a pollutant after weather normalization.

East Asian winter monsoon index

In this study, the EAWN index proposed by Jeong and Park (2017) is used, which reflects monsoonal variations in meteorological variables such as surface air temperature, precipitation, and circulation. The definition of the EAWM index is as follows:

$$I_{EAWM} = \frac{(2 \times SLP_1^* - SLP_2^* - SLP_3^*)}{2}$$

Where SLP_1^*, SLP_2^*, and SLP_3^* indicate the normalized area-averaged SLP over Siberia (40°-60°N, 70°-120°E), the North Pacific (30°-50°N，140°E-170°W), and the Maritime continent (20°S-10°N, 110°-160°E), respectively. The EAWM indices were calculated by using the JRA55 (the 55-year Japanese Reanalysis Project data set) data.

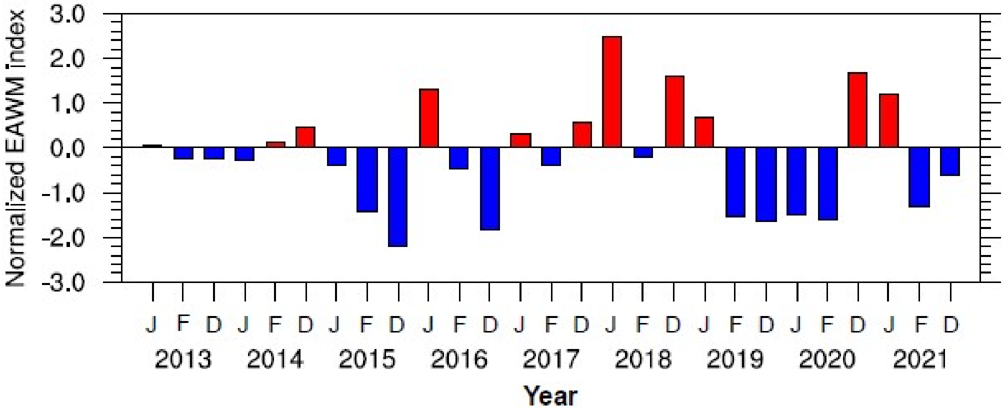


Fig.S3. Normalized time series of mean EAWM indices from 2013 to 2021.

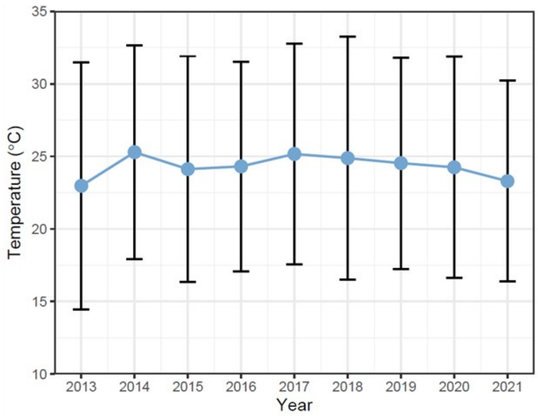


Fig.S4. Temperature variation from 2013 to 2021 at SDZ (blue circle is the average temperature and horizontal line represents ±1 σ).

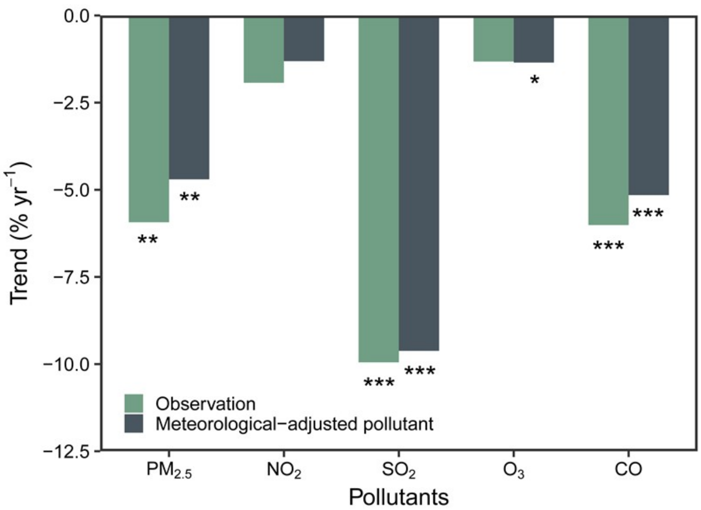
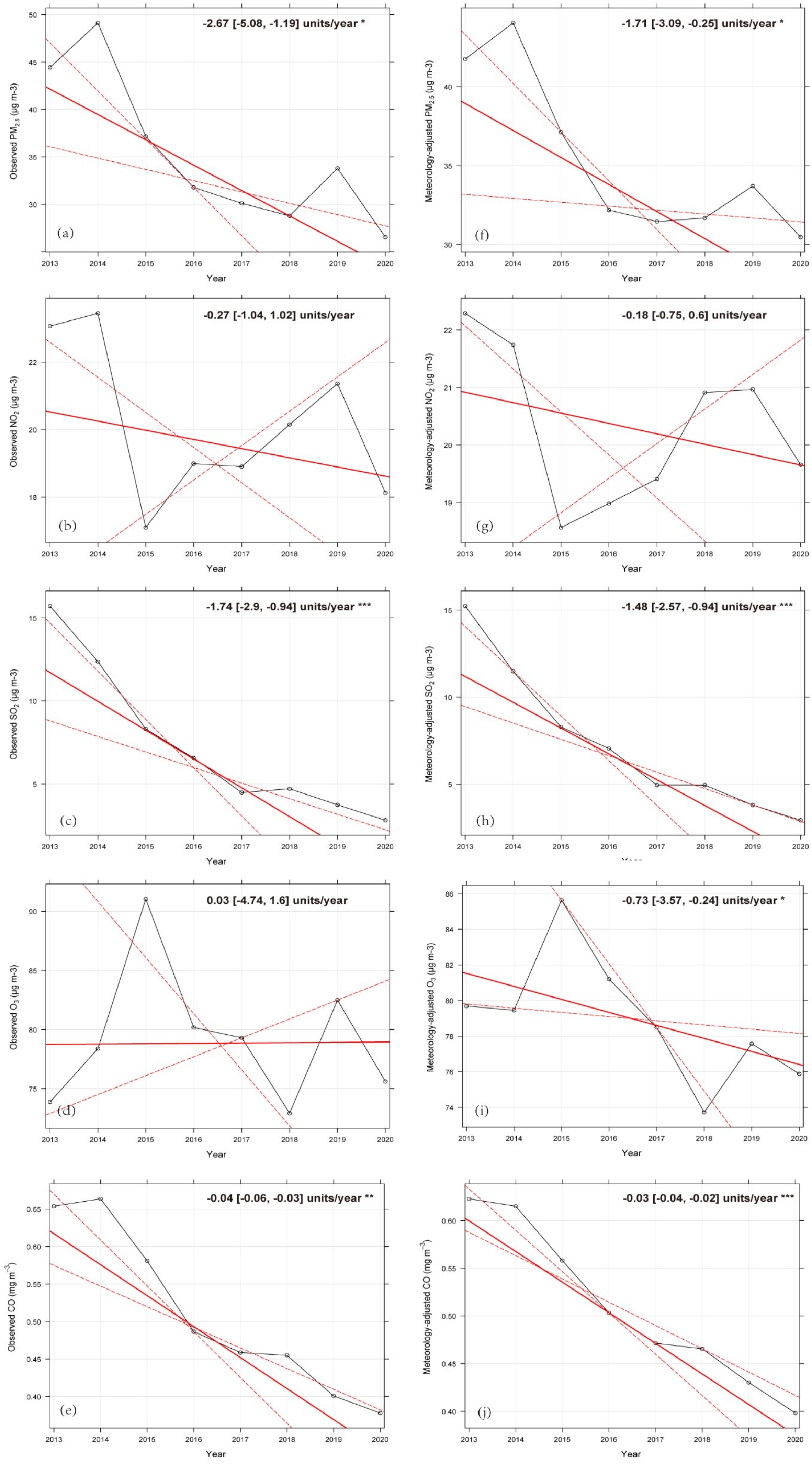


Fig.S5. Mean annual change rates from 2013 to 2021 (*, **, and *** represent p-values of 0.1, 0.01, and 0.001, respectively).



-2.67 [-5.08, -1.19] units/year *
Observed PM2.5 (μg m-3)
(a)
Year
-1.71 [-3.09, -0.25] units/year *
Meteorology-adjusted PM2.5 (μg m-3)
(f)
-0.27 [-1.04, 1.02] units/year
Observed NO2 (μg m-3)
(b)
-0.18 [-0.75, 0.6] units/year
Meteorology-adjusted NO2 (μg m-3)
(g)
-1.74 [-2.9, -0.94] units/year ***
Observed SO2 (μg m-3)
(c)
-1.48 [-2.57, -0.94] units/year ***
Meteorology-adjusted SO2 (μg m-3)
(h)
0.03 [-4.74, 1.6] units/year
Observed O3 (μg m-3)
(d)
-0.73 [-3.57, -0.24] units/year *
Meteorology-adjusted O3 (μg m-3)
(i)
-0.04 [-0.06, -0.03] units/year **
Observed CO (mg m-3)
(e)
-0.03 [-0.04, -0.02] units/year ***
Meteorology-adjusted CO (mg m-3)
(j)

Fig.S6 Trends of annual average air quality parameters before (a, b, c, d, and e) and after (f, g, h, i, and j) normalization of weather condition. The red line represents the Theil-Sen trend, the dashed red lines show the 95% confidence intervals and the *** shows that the trend is significant to the 0.001 level.

Table S1. RF model performance for testing dataset with all statistical metrics (in hourly resolution)

Pollutants	**FAC2**	**MB**	**MGE**	**NMB**	**NMGE**	**RMSE**	**r**	**COE**	**IOA**
$PM_{2.5}$	0.82	0.24 μg/m^3	10.30 μg/m^3	0.01	0.30	16.50 μg/m^3	0.90	0.58	0.80
NO_2	0.86	0.15 μg/m^3	5.37 μg/m^3	0.00	0.27	8.33 μg/m^3	0.89	0.58	0.80
SO_2	0.77	0.13 μg/m^3	2.40 μg/m^3	0.02	0.34	4.86 μg/m^3	0.93	0.65	0.83
O_3	0.93	0.21 μg/m^3	13.30 μg/m^3	0.00	0.17	19.4 μg/m^3	0.93	0.66	0.83
CO	0.95	0.01 mg/m^3	0.11 mg/m^3	0.01	0.23	0.19 mg/m^3	0.92	0.62	0.81

Table S2. The relative changes (%) of pollutants from 2013 to 2017, and 2013 to 2020

	Observation						**Meteorology-adjusted**				
	$PM_{2.5}$	**NO_2**	**SO_2**	**O_3**	**CO**		**$PM_{2.5}$**	**NO_2**	**SO_2**	**O_3**	**CO**
2013-2017	-32.2	-18.2	-71.5	7.3	-29.2		-24.6	-13.0	-67.8	-1.5	-24.2
2013-2020	-40.3	-21.6	-82.1	2.3	-41.5		-27.0	-11.7	-80.9	-4.8	-35.5

References:

Jeong, J.I., Park, R.J., 2017. Winter monsoon variability and its impact on aerosol concentrations in East Asia. Environmental Pollution 221, 285-292.