



Supplement of

Measurement report: Long-term variations in surface NO_x and SO_2 mixing ratios from 2006 to 2016 at a background site in the Yangtze River Delta region, China

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The levels of NO, NO₂ and NO_x.

As shown in the Tab. S1, the average annual mixing ratio of NO, NO₂ and NO_x at LAN over the 11 years is 1.1 ppb, 12.5 ppb and 13.6 ppb, respectively. The average ratio of NO₂/NO_x is 0.92, which indicates that NO₂ accounts for a huge percentage of NO_x and is dominated by ageing air masses locally. It can also be seen from Table S1 that the annual average NO₂ mixing ratio follows roughly the same trend as NO_x, so we have discussed the long-term NO_x variations in the manuscript and long-term NO₂ variations in the supplement.

Table S1. Statistics of NO, NO₂ and NO_x levels from 2006 to 2016 at LAN.

year	NO	NO ₂	NO _x	NO ₂ /NO _x
2006	0.8	12.1	12.9	0.94
2007	1.1	12.7	13.8	0.92
2008	1.0	12.0	13.0	0.92
2009	1.0	12.1	13.1	0.92
2010	1.6	12.5	14.1	0.87
2011	1.3	14.1	15.4	0.92
2012	1.6	13.8	15.4	0.90
2013	1.0	13.5	14.5	0.93
2014	0.8	12.1	12.9	0.94
2015	1.0	11.0	12.0	0.92
2016	1.1	11.1	12.2	0.91
average	1.1	12.5	13.6	0.92

Monthly average NO₂ and NO mixing ratios.

As shown in the Fig. S1, the average seasonal variation in NO₂ and NO is similar, with highest level in December (20.3 ± 3.6 ppb and 3.1 ± 1.2 ppb) and lowest in July (6.5 ± 0.6 ppb) and June (0.5 ± 0.2 ppb), respectively.

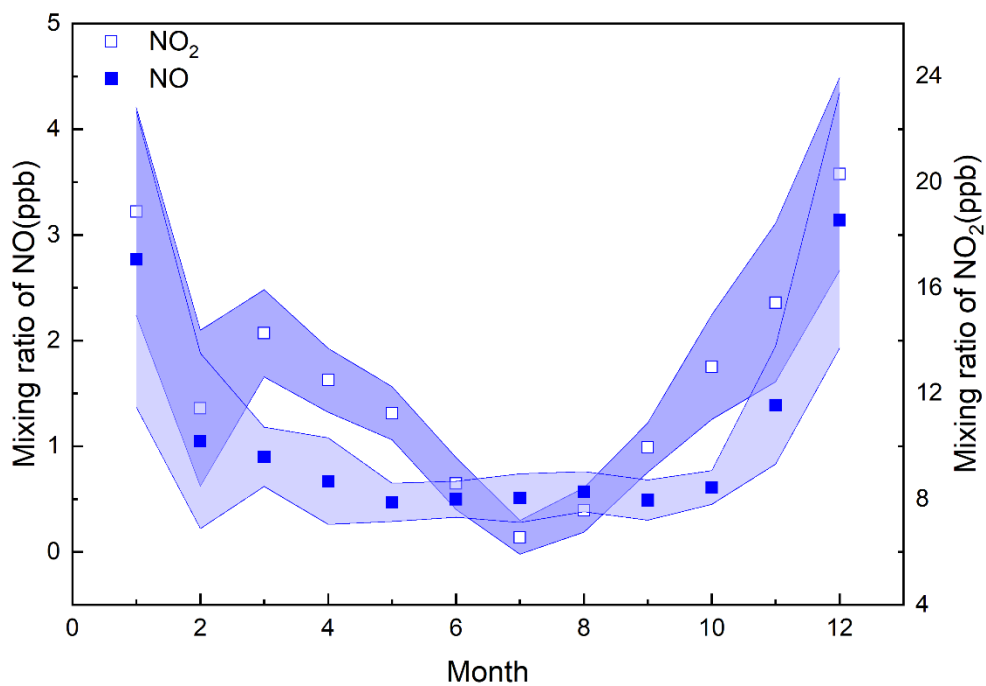


Figure S1: Monthly average NO₂ and NO mixing ratios at LAN. The shadows represent 1 standard deviation calculated from monthly means.

Diurnal variations in NO₂ and NO mixing ratios.

As shown in Fig.S2, an evident NO peak at 9:00 am during morning rush hours indicates the presence of fresh emissions mainly from vehicle emissions.

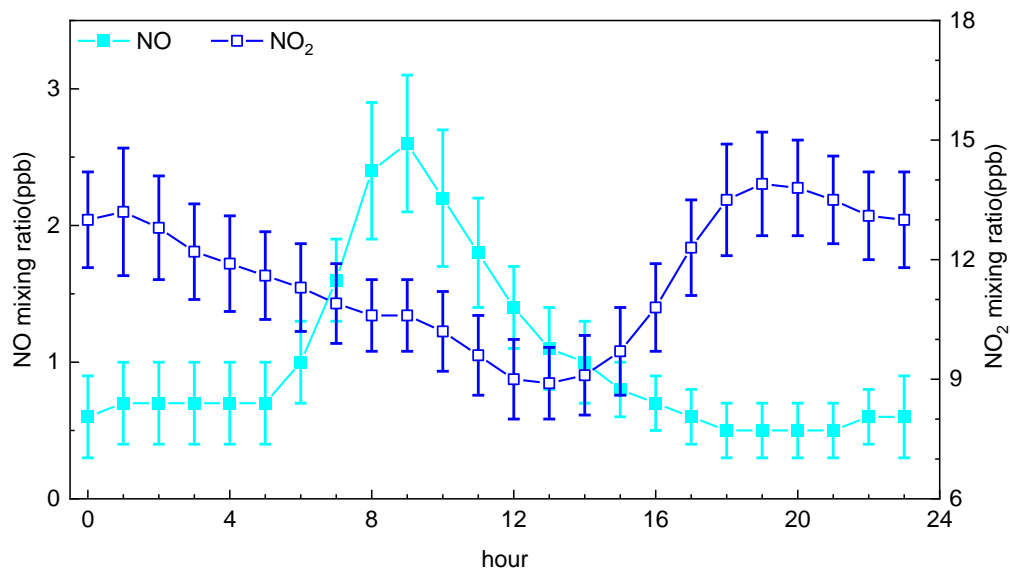


Figure S2: Annual average diurnal variations in NO (left axis) and in NO₂ (right axis) at LAN.

Annual mean NO₂ mixing ratio at LAN compared with OMI NO₂ vertical column density over YRD.

We compared the long-term trends in LAN NO₂ mixing ratio with OMI NO₂ vertical column density (Fig.S3a) and found significant correlations between the two (Fig.S3b, $P < 0.01$), which indicate a good regional representativeness of LAN measurements. In the Fig.S3a, the percentage changes of NO₂ in LAN and OMI are +2.23%/yr ($P = 0.17$) and +5.87%/yr ($P < 0.01$) from 2006 to 2011 (based on 2006), respectively. And the percentage changes of the two are -4.98%/yr ($P < 0.01$) and -4.22%/yr ($P < 0.01$) from 2011 to 2016 (based on 2011), respectively.

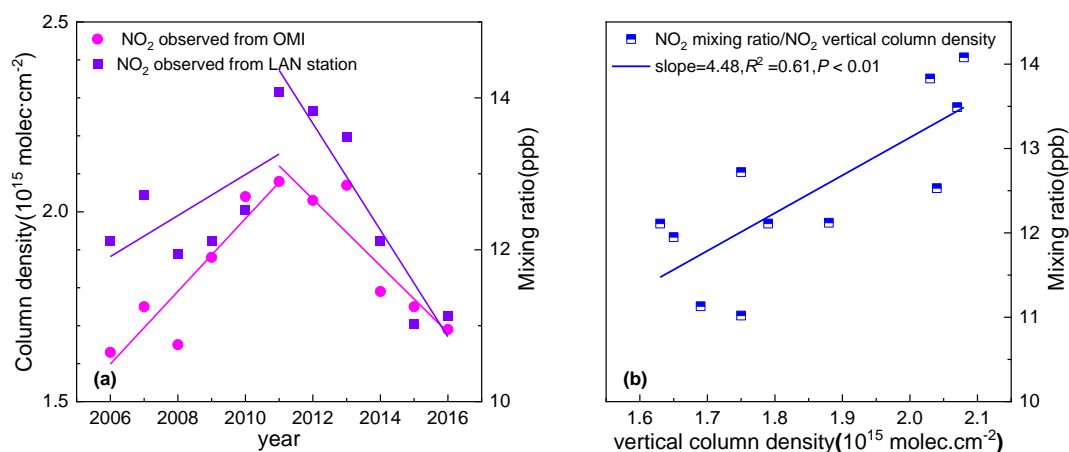


Figure S3: Annual mean NO₂ mixing ratio at LAN (a, right axis) compared with OMI NO₂ vertical column density over YRD (a, left axis) and the linear fit of both (b).