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## Comment on acp-2021-227

Qingqing Yin et al.

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Author comment on "Measurement report: Long-term variations in surface NO<sub>x</sub> and SO<sub>2</sub> mixing ratios from 2006 to 2016 at a background site in the Yangtze River Delta region, China" by Qingqing Yin et al., Atmos. Chem. Phys. Discuss.,  
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We thank for the constructive comments and suggestions. We revised our manuscript according to the comments and suggestions. The following list the point-to-point response to the comments. The changed texts were highlighted with yellow color.

### Response to comments by referee # 1

General comments:

This paper reports on NO<sub>x</sub> and SO<sub>2</sub> measurements at the background site in the Yangtze River Delta region in China.

The site, the instrumental setup, quality control and the data processing procedures have been described in detail. Data are compared to other data from other measurement sites. The long-term trend of both SO<sub>2</sub> and NO<sub>x</sub>, their diurnal and seasonal behavior are discussed and compared to emission data.

There are only few NO<sub>x</sub> and SO<sub>2</sub> datasets on background sites published and analyzed in depth so far so I would recommend this paper being accepted for publication after the following questions are answered.

Specific comments:

- Line 85 In the information and methods part, the instrumental setup is described. Here, an essential part is missing. The method to convert NO<sub>2</sub> into NO for detection should be given, as well as the method for determining the conversion efficiency. Has gas phase titration been used? Also, it should be mentioned if data were corrected for humidity and ozone effects.

Response: In Model 42C-TL trace-level chemiluminescent analyzer, NO<sub>2</sub> is converted to NO by a molybdenum NO<sub>2</sub>-to-NO converter heated to about 325°C. The converter efficiency was checked annually using gas phase titration (GPT). If the converter efficiency is less than 96%, replace the converter. We add the information in the revised paper. Please see page 3, line 89 for the revision.

- Line 135 One major concern is that the paper describes trends and seasonal behavior of NO<sub>x</sub>. However, of the nitrogen oxides, NO<sub>2</sub> has the major impact on health. NO<sub>2</sub>

data should be included into table 1 and discussed. What is the long-term trend of NO<sub>2</sub>?

Response: In this regional background station, NO<sub>2</sub> was the dominant form of NO<sub>x</sub>, accounting for 82.2 % of NO<sub>x</sub>, so we didn't present the trends and seasonal behavior of NO<sub>2</sub>. In the revised manuscript, we included the NO<sub>2</sub> in Table 1 and text, but trends and seasonal behavior of NO<sub>2</sub> in supplementary material.

- Line 167 As the authors point out satellite observations are a valuable tool when analyzing station data. How does the long-term trend of the OMI NO<sub>2</sub> observation compare to the NO<sub>x</sub> data at the site? Likewise, comparison of in situ observation with station data could help to differentiate between boundary layer effects and emission effects when discussing the diurnal behavior of NO<sub>x</sub>. How does the diurnal behavior of the satellite observation compare to the diurnal behavior of station data?

Response: Thanks for the suggestion. We think it would be a good idea to compare in situ observation with satellite observations to distinguish between boundary layer effects and emission effects. But, given our current level of knowledge, we need to learn further to complete the work. Here, we compare the monthly average satellite products for NO<sub>2</sub> with the monthly mean surface NO<sub>2</sub>, since OMI covers a point on the ground about once every two days.

- Line 207 The pollution roses in Figure 5 show that SO<sub>2</sub> and NO<sub>x</sub> mixing ratios depend not only on the windspeed but also on the wind direction. Is it possible to add a plot to figure 5 which shows the dependency on wind direction?

Response: Thanks for the suggestion. Rose maps are often used to discuss the relationship between wind direction and pollutant concentration, which details had been plotted in Figure 6 and analyzed in section 3.4.

- Line 220 Changes in relative humidity can often be explained with changes in airmasses which are advected from different sites. How does relative humidity change with wind direction? Can the wind direction explain the change of NO<sub>x</sub> and SO<sub>2</sub> with changing relative humidity?

Response: Thanks for your suggestion. We plot the relative humidity change with wind direction together with NO<sub>x</sub> and SO<sub>2</sub> rose maps (Figure R1). Also comparing them with figure 1, we think it cannot explain the change of NO<sub>x</sub> and SO<sub>2</sub> with changing relative humidity.

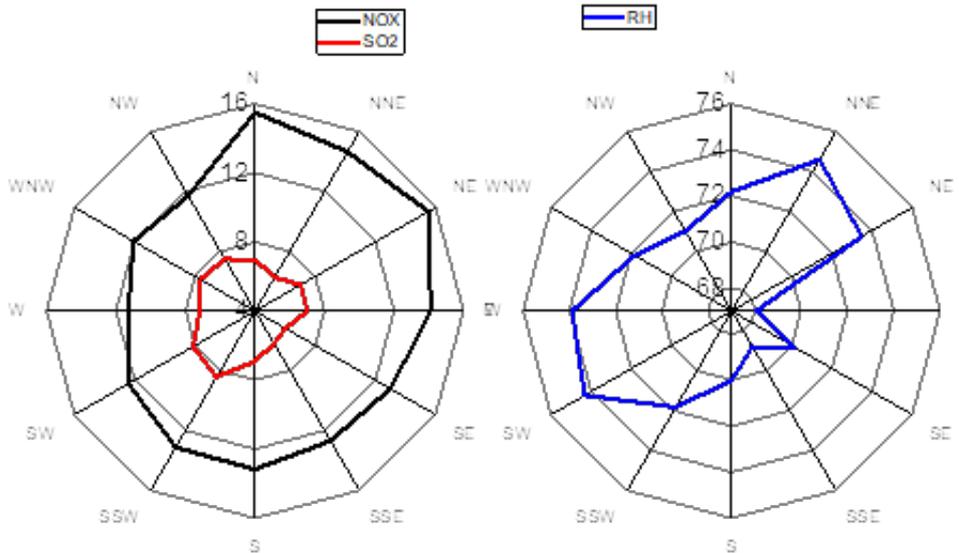


Figure R1. NO<sub>x</sub>, SO<sub>2</sub>, and RH rose maps

- Line 237 The authors write that the main source of SO<sub>2</sub> and NO<sub>x</sub> are east from the site as the show it in figure 7. However, in figure 6 it can be observed that highest mixing ratios were measured with wind coming from west. How can this discrepancy be explained?

Response: Thanks. The wind concentration rose diagram (Figure 6) is a method that can effectively identify short-range transport sources near the ground, while the CWT method (Figure 7) can effectively identify long-range transport sources. Based on a comparison of the two, it can be seen that from the perspective of local emissions, atmospheric NO<sub>x</sub> and SO<sub>2</sub> at the LAN station are mainly from the northeast and southwest of the station, while long-range transport is influenced by the east.

- Line 305 The diurnal behavior is already discussed in chapter 3.3. I would suggest merging chapter 3.3 with lines 305 to 341.

Response: Thanks for the suggestion. Chapter 3.3 aims to provide an overview of the characteristics of the diurnal behavior of NO<sub>x</sub> and SO<sub>2</sub> over the observation period, while lines 305 to 341 focus on their **long-term** characteristics and causes. So, we kept to separate chapter 3.3 from lines 305 to 341.

- Line 327 If the disappearance of the NO<sub>x</sub> peak at 1:00 A.M. were due to reduction of industrial emissions, why should industrial emissions peak at 1:00 A.M.? Shouldn't the effect be seen all over the night?

Response: Yes, NO<sub>x</sub> peak at 1:00 AM also puzzled us. A small peak in NO<sub>x</sub> and SO<sub>2</sub> occurred between 01:00 and 02:00, which might be related to nighttime emissions from unscrupulous enterprises (Fan et al., 2013) or the lower electricity prices after midnight in response to the financial pressure of the 2008 economic crisis and the corresponding increase in electricity prices for industrial users (Sun, 2008). But it's really hard to tell exactly why these small peaks dominate after midnight. These two causes also feel too speculative, so we just present the result here.

We revised the sentences as (Page 11, line 334):

The disappearance of the small peak around 01:00 at night during 2012–2016 may be related to the introduction of stricter air pollution control policies for factories that emit at night. Small peaks in NO<sub>x</sub> and SO<sub>2</sub> occurred between 01:00 and 02:00, which might be related to nighttime emissions from unscrupulous enterprises (Fan et al., 2013) or more production activities with lower electricity prices after midnight in response to the financial pressure of the 2008 economic crisis and the corresponding increase in electricity prices for industrial users (Sun, 2008). In spite of these two reasons, however, it's really hard to tell exactly why these small peaks dominate after midnight.

Fan, Y., Fan, S., Zhang, H., Zu, F., Meng, Q., and He, J.: Characteristics of SO<sub>2</sub>, NO<sub>2</sub>, O<sub>3</sub> volume fractions and their relationship with weather conditions at Linan in summer and winter, *J. Atmos. Sci.*, 121–128, 2013.

Sun, W.: "A two-pronged approach of "limiting coal prices" and "raising electricity prices, Yangtze River Delta, 58–60, 2008.

- Line 337 To my knowledge, the impact of traffic on SO<sub>2</sub> emission in China is of minor importance. Have you considered residential sources, which are after industrial emissions and power plant emissions the third most important source of SO<sub>2</sub> according to the Multi-resolution Emission Inventory for China (MEIC)?

Response: Thanks. You are right. For the evening peaks, the residential sources should be important for SO<sub>2</sub> because it's also in cooking hours. We revised sentence as: "The formation of the evening peaks of NO<sub>x</sub> and SO<sub>2</sub> may be mainly related to the increase in motor vehicle and residential sources emissions, which are stronger in the rush and cooking hours and that of SO<sub>2</sub> may be probably more due to the reduction of power plants emissions." Please see Page 12, line 348 for the revision.

- Line 369 Data availability: A link should be provided to where the data are stored in the GAW archive.

Response: As far as I know, the data of NO<sub>x</sub> and SO<sub>2</sub> data for this site are not available in the GAW archive. The specific reasons are complex.

Technical corrections:

- Title: Measurement report: Long-term variations in surface NO<sub>x</sub> and SO<sub>2</sub> from 2006 to 2016 at a background site in the Yangtze River Delta region, China

Better:

Title: Measurement report: Long-term variations in surface NO<sub>x</sub> and SO<sub>2</sub> mixing ratios from 2006 to 2016 at a background site in the Yangtze River Delta region, China

Response: Accepted. Please see page 1, line 1 for the revision.

- Line 185: The seasonal average diurnal variation in NO<sub>x</sub> showed a morning peak of NO<sub>x</sub> in summer at 08:00, which is 1 to 2 h earlier than during other seasons (Fig. 4c). This sentence is not clear to me. What is it compared to?

Response: We revised the sentence. "In summer, the seasonal average diurnal variation in NO<sub>x</sub> showed a morning peak at 08:00, which time is 1 to 2 h earlier than that occurred in other seasons (Fig. 4c)." Please see Page 7, line 192 for the revision.

- Line 221: different periods are well consistent

Response: Accepted. Please see Page 7, line 224 for the revision.

- Line 222: A blank is missing

Response: Accepted. Please see Page 8, line 225 for the revision.

- Line 250: Please give a reference for the Ecological and Environmental Status Bulletin.

Response: Accepted. Please see Page 8, line 225 for the revision.

- Line 254: smaller than those

Response: Accepted. Please see Page 9, line 258 for the revision.

- Figure 5: The dependencies of SO<sub>2</sub> on meteorological parameters in the figure is blurred from the underlying trend. In figure 5h it cannot be seen if data for 2014-2016 change at all. Maybe it is better to plot changes relative to a mean value.

Response: Accepted. Please see Page 24, figure 5 for the revision.

Figure 6: I would suggest using the same color for NO<sub>x</sub> in all the seasons in this plot and label the plots instead.

Response: Accepted. Please see Page 25, figure 6 for the revision.

- Figure 11: The different y scales in Figures 11a to Figures 11c makes comparison between the periods difficult. I would suggest using the same scale.

Response: Accepted. Please see Page 29, figure 11 for the revision.

- Figure 11: I would suggest naming the periods in the figure caption.

Response: Accepted. Please see Page 29, figure 11 for the revision.

## **Response to comments by referee # 2**

### General comments

The authors present a concise study on long-term trends of NO<sub>x</sub> and SO<sub>2</sub> in the region of the Yangtze River Delta based on data acquired at the global GAW station Lin'an.

They interpret diurnal, seasonal and long-term changes and put them in context with trends measured in several other regions or cities in China and across the globe.

They assess correlations with meteorological parameters and emissions by different sources.

To identify source regions a potential source analysis of NO<sub>x</sub> and SO<sub>2</sub> is applied on the YRD region.

Changes in average diurnal patterns of NO<sub>x</sub> and SO<sub>2</sub> from 2006 to 2016 were found and attributed to long-term changes of vehicle emissions and industrial emissions, respectively.

#### Specific comments

I.30, I.307, table 1: Why not denoting it SO<sub>2</sub>/NO<sub>x</sub> or Sulph./Nitrog.? S/N might be mixed up with signal-to-noise ratio.

Response: Accepted. We use SO<sub>2</sub>/NO<sub>x</sub> instead of S/N in the revised version.

I.85: For NO<sub>x</sub>: The method of NO<sub>2</sub> to NO conversion is missing and must be explained. Furthermore, were corrections for humidity (respective quenching for CLD technique) and ozone reaction within the inlet line applied on the presented data?

Response: In model 42C-TL trace-level chemiluminescent analyzer, NO<sub>2</sub> is converted to NO by a molybdenum NO<sub>2</sub>-to-NO converter heated to about 325°C. The converter efficiency was checked annually using gas phase titration (GPT). If the converter efficiency is less than 96%, replace the converter. We add the information in the revised paper. Please see Page 3, line 89 for the revision.

I.97: Item (4) might describe more specifically what kind of checks, testings are done and what self-diagnosis does comprise of or even better give a reference where it is explained in more detail. For example, I'm wondering what self-diagnosis means: Is it applying internal thresholds for operating parameters and, if yes, which one and what are the consequences for the instrument or measurements? Is this automatically done by the instrument or by the data acquisition system?

Response: Mostly, the instrument self-diagnosis is applying internal thresholds for operating parameters, which can alert people to carry out manual testing, checking, and maintenance on the instrument. It can't be described in a few words. Technical report from U.S. Environmental Protection Agency can be a reference here. We add a reference of US EPA (2017) in the text.

US EPA Quality Assurance Handbook for Air Pollution Measurement Systems, Volume II, Ambient Air Quality Monitoring Program, EPA-454/B-17-001, 2017.

I.215: Is it adequate to call it already "effect" if this is an investigation of correlations? Is causality approved yet?

Response: Thanks. We have corrected "effect" to "correlation". Please see Page 8, line 218 for the revision.

I.328: Reference for a change of (specific) air pollution control measures?

Response: You might mean what specific air pollution control measures causing the disappearance of the NO<sub>x</sub> peak at 1:00 A.M? PLS see our response to referee #1, question 8.

#### Technical corrections

Figure 2: P (hPa), factor 10 missing?

Response: Thanks. The factor of 10 is missing. The unit is kPa. Please see Page 21, Figure 2 for the revision.

