Response to the Editor's Comments:

As discussed by the two referees, this paper presents a useful analysis of particulate nitrate pollution in the North China Plain. I concur in their judgment. In addition to the comments of those referees, I suggest that two additional points be discussed:

Response: we thank the editor for handling and evaluating our submission. These comments are very helpful and we have revised the manuscript according to these comments. Below are the responses to the specific comments, with the changes in the manuscript highlighted in red color.

1) In the Abstract the authors note: "The nitrate/$PM_{2.5}$ and nitrate/sulfate ratios have significantly increased in Ji'nan (2005-2015) and at Mt. Tai (from 2007 to 2014), indicating the worsening situation of regional nitrate pollution." And likewise "This study provides observational evidence of rising trend of nitrate aerosol …" These statements are necessarily correct only if the absolute concentrations of $PM_{2.5}$ and sulfate have remained constant (or increased). If these two species in the denominator of the two ratios have decreased more rapidly than the ratios themselves, then the regional nitrate pollution may be improving in an absolute (but not relative) sense. A short discussion and clarification of this issue should be included.

Response: we explored the trends in the absolute concentrations of $PM_{2.5}$, sulfate and nitrate in Ji'nan and at Mt. Tai. The figures are shown below. Indeed, the ambient levels of $PM_{2.5}$ and sulfate have rapidly decreased in the NCP region over the past decade, largely owing to the stringent control of SO_2 emissions and primary particles. In comparison, the absolute concentrations of nitrate in $PM_{2.5}$ showed an increasing trend from 2005 (or 2007) to 2015 (0.29 and 0.39 $\mu g/m^3/yr$). This confirms the increasing trend of nitrate aerosol pollution in this region. Nevertheless, the available observations since 2011 also showed a decrease in the absolute levels of nitrate aerosol in Ji'nan. This trend may be true given the strict NOx emission control by the central government of China since 2011, but it may be also interfered by the higher aerosol pollution observed during the campaign of 2011 that should be due to the unfavorable meteorological conditions. Anyway, more measurement studies are required to further examine the recent trend of nitrate aerosol since 2011 and assess the impact of the NOx control implemented by the government. The following figures and discussion have been added to clarify this issue in the revised manuscript.

"We also examined the trends in the absolute concentrations of $PM_{2.5}$, nitrate and sulfate in urban Ji'nan and at Mt. Tai (see Fig. S2). As expected, the ambient concentrations of $PM_{2.5}$ (6.3 and 1.4 $\mu g\ m^{-3}\ yr^{-1}$) and SO_4^{2-} (2.1 and 1.2 $\mu g\ m^{-3}\ yr^{-1}$) have rapidly decreased at both locations during the past decade, which should be largely attributed to the stringent control of SO_2 emissions and primary particles. In comparison, the absolute concentrations of NO_3^- showed an increasing trend with average rates of change of 0.39 and 0.29 $\mu g\ m^{-3}\ yr^{-1}$. This confirms the increase of nitrate aerosol pollution in the NCP region. Nevertheless, the available observations since 2011 also showed a decrease in the absolute levels of nitrate aerosol in Ji'nan. This trend may be true considering the strict NOx emission control of China since 2011, but it may be also partly interfered by the higher aerosol pollution observed

during the campaign of 2011 with unfavorable meteorological conditions. More measurement efforts are urgently needed to further examine the recent trend of nitrate aerosol after 2011 and evaluate the impact of the NOx emission control implemented by the Chinese government."

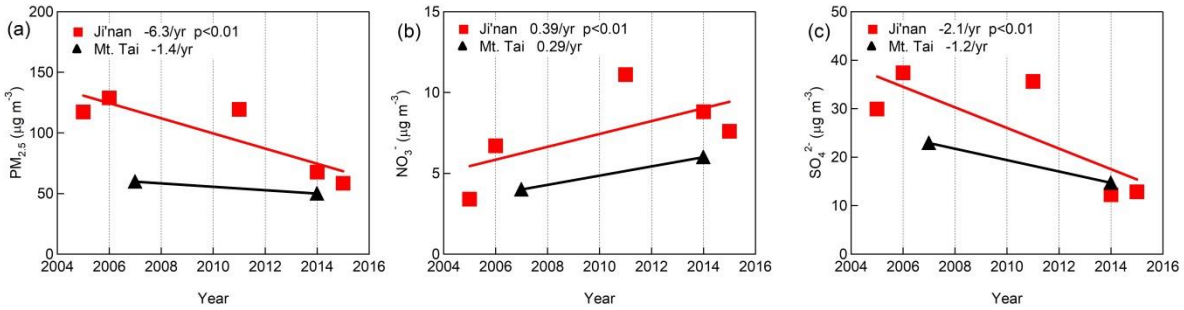

Figure S2. Long-term trends in the absolute concentrations of (a) $PM_{2.5}$, (b) NO_3^-, and (c) SO_4^{2-} in urban Ji'nan and at Mt. Tai in summertime from 2005 to 2015. The fitted lines are derived from the least square linear regression analysis, with the slopes and p values (99% confidence intervals) denoted.

2) In the Conclusions the authors "recommend that further reduction of anthropogenic emissions of NO_X should be the most efficient pathway for the current control of nitrate aerosol …" The data discussed in the paper were collected in 2014 and earlier years. Satellite data (e.g., Liu et al., 2017) suggest that NO_x over the North China Plain was increasing during the period covered by these data, but has been decreasing rapidly since 2014. The authors should briefly discuss the likely impact of this NO_x reduction.

Response: indeed, some very recent studies have indicated the decrease in the anthropogenic emissions and ambient abundances of NOx over eastern China in the past five years. It is definitely expected that such reduction of NOx would contribute to a decrease in the fine nitrate aerosol in this region. Nevertheless, this still needs to be further confirmed by the long-term observations in the near future. The following discussion has been added in the revised manuscript.

"Some recent studies have reported the rapid decrease in the NOx abundances over eastern China since 2011 (Liu et al., 2017). It can be expected that such reduction of NOx would help to alleviate the nitrate particulate pollution in China. More observational studies are needed to further examine the trend in the nitrate aerosol and assess the contributions of the strict NOx control of China."

Reference:

Liu, F., Beirle, S., Zhang, Q., van der A, R. J., Zheng, B., Tong, D., & He, K. (2017). NO_x emission trends over Chinese cities estimated from OMI observations during 2005 to 2015. Atmospheric Chemistry and Physics, 17, 9261–9275.