Responses to Reviewers:

Reviewer 1:

We thank the reviewer for the careful reading of the manuscript and helpful comments. We have revised the manuscript following the suggestions as is described below.

(1) This manuscript apparently focused on the daytime O3 formation mechanisms (VOC-limited or NOx-limited). But most of the analysis is conducted for daily mean O3 concentration. Daytime O3 and nighttime O3 are affected by totally different processes/mechanisms and they may have different variations/trends. The tread of daily mean O3 discussed mostly in this manuscript may be dominated by the trend of nighttime O3, which are not governed by the daytime O3 formation mechanisms (VOC-limited or NOx-limited). **Given that this manuscript wants to focus on NOx-limited or VOC-limited, I would recommend the authors change their analysis to focus on daily maximum O3.**

According to the excellent suggestion of the reviewer, we analyzed the variation of daily maximum O_3 concentration instead of daily mean O_3 concentration in Sect. 3.1. The new results were re-plotted in Fig. 2. It was showed that the annual variation of daily maximum O_3 concentration also presented significant increasing trend in XJH and PD sites which was same as that of daily mean O_3 concentration. The increasing rate was 0.808 ppbv.yr^{-1} at XJH site, which was lower than that at PD site of 1.374 ppbv.yr^{-1}. The above new results were included in the revised version. Furthermore, we calculated the variability of daily maximum 8h-O_3 concentration, which also exhibited the same increasing trends at the rate of 1.063 and 1.403 ppbv.yr^{-1} at XJH and PD sites respectively.

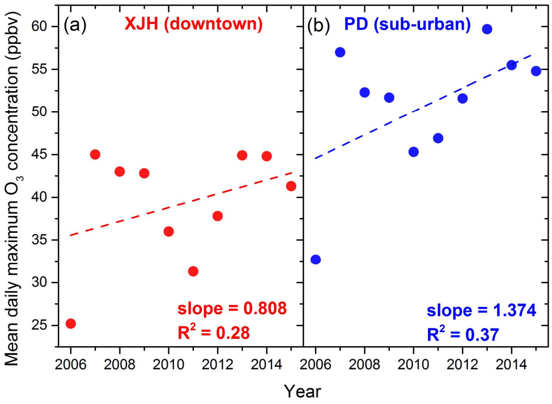

Fig. 2, Annual variation of daily maximum O_3 concentration during 2006 to 2015 at (a) XJH and (b) PD respectively.

(2) Showing change of mean diurnal variation of O3 would be helpful to identify the different trends of daytime/nighttime O3. In the few places when daytime/nighttime O3 are separately discussed in this manuscript, the time ranges for the “daytime”/”nighttime” are not specified. Thus, it is hard for this reviewer to judge whether the “daytime” O3 is only affected by O3 formation mechanisms or the “daytime” O3 is still affected by the O3 removal processes during nighttime and early morning. Thus, showing the trend of mean diurnal variation is critical.

Thanks for the reviewer's suggestion. We calculate the variability of hourly O_3 concentration during 2006 to 2015 to investigate the diurnal variations of O_3 trend in Sect. 3.4. The O_3 concentration showed increasing trend both in daytime (8:00-18:00, LST) and nighttime (19:00-07:00, LST) at XJH and PD sites. The nighttime O_3 increased more significantly than daytime O_3 at XJH, with the increasing rate of 1.239 and 0.956 ppbv.yr^{-1} respectively. While at PD the O_3 concentration increased by 1.338 ppbv.yr^{-1} in daytime which was higher than that in nighttime of 1.028 ppbv.yr^{-1}. In comparison, nighttime O_3 presented higher increasing rate at downtown site XJH than that at sub-urban site PD due to more NO emissions at urban center or the enhanced urban effects (Hu et al., 2013). The new results were re-plotted in Fig. 6. In addition, we also compared the seasonal variability of daytime and nighttime O_3 concentrations at XJH. The larger O_3 variability in nighttime than daytime was observed in spring, summer and autumn. For example, the nighttime O_3 concentration increased at 1.341, 1.159 and 1.525 ppbv yr-1 in spring, summer and autumn respectively, which are more significant than that of 1.008, 0.378 and 1.370 ppbv yr-1 in daytime. The variability of winter O_3 concentrations in daytime and nighttime are generally close perhaps due to the lower O3 photochemical productions. The seasonal results were plotted in the Fig. 7.

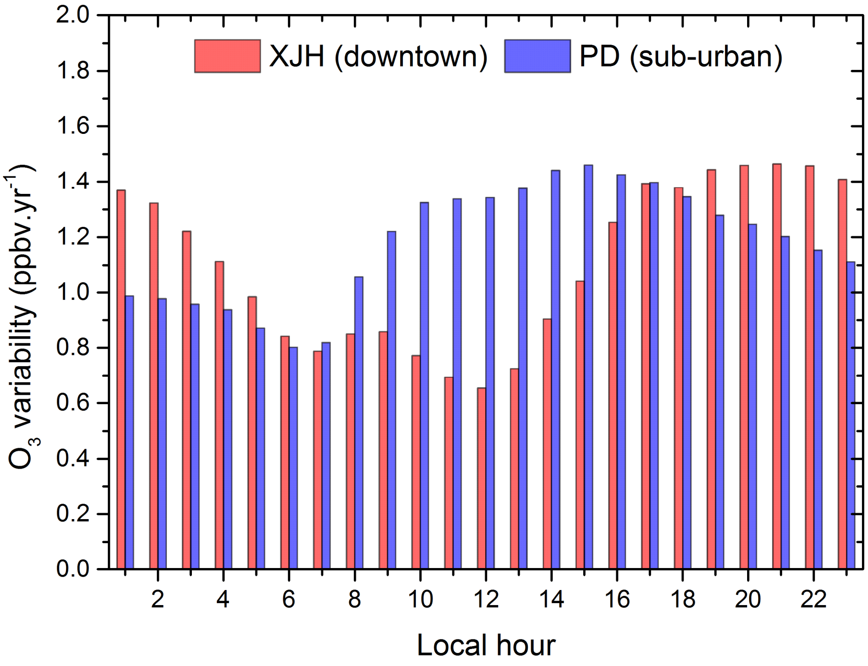

Fig.6 The variability of hourly O_3 concentration during 2006 to 2015 at XJH and PD respectively

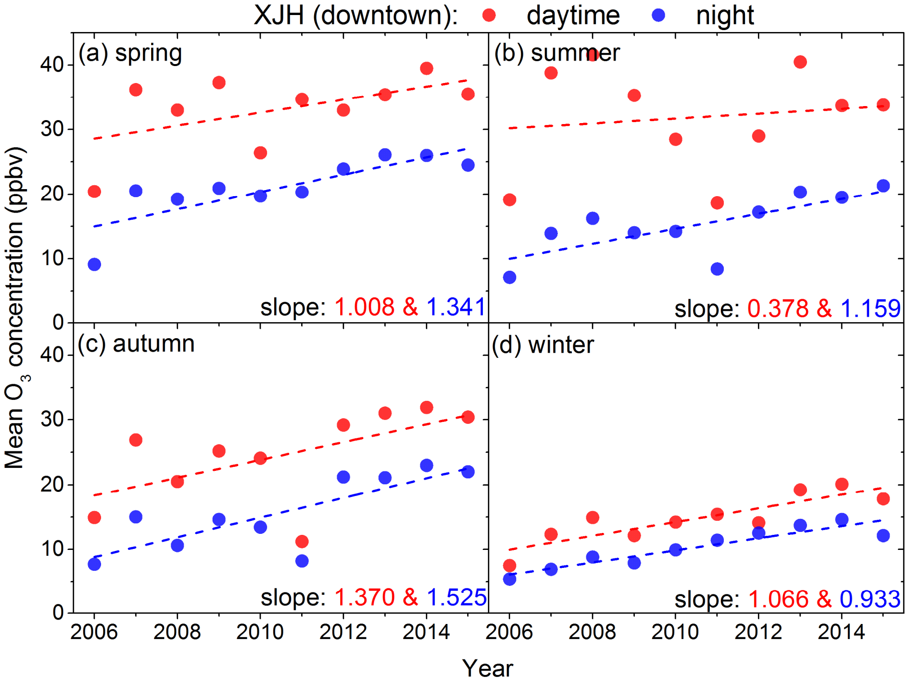

Fig.7 The variability of daytime (08:00-18:00, LST) and nighttime (19:00-07:00, LST) O_3 concentration during 2006 to 2015 at XJH in (a) spring, (b) summer, (c) autumn, (d) winter respectively.

(3) The current writing sounds like nighttime O3 is only affected by NO titration. Actually nighttime O3 is affected by three main processes, i.e., NO titration, dry deposition and vertical mixing (Hu et al., 2013). Both of the latter two processes are related to nighttime turbulence, which are further related to extent of urbanization. Thus the increasing trend of nighttime O3 may reflect reduced NO titration, as well as enhanced nighttime vertical mixing, or say less stable nighttime boundary layer, which may be induced by enhanced urban effects through the years (Hu et al., 2016).

Thank for this comment. We agreed that both dry deposition and nighttime turbulence influenced the nighttime O_3 concentration according to the work by Hu et al. (2013). We checked the vertical temperature gradient between 1000 hpa and 950 hpa at 20:00 (LST) in Shanghai to indicate the nighttime turbulence intensity based on sounding data, while presented no significant trend during 2010 to 2015. Furthermore, the PBL height at 20:00 (LST) retrieved from MPL measurements also varied insignificantly (slight decreasing trend) during the same period. Based on the above measurements, the variation of turbulence at night may have only minor contribution to the nighttime O_3 increasing in Shanghai. However the effect of dry deposition could not be excluded by lacking of measurements, which need further investigation. Such discussion has been included in Sect. 3.4.

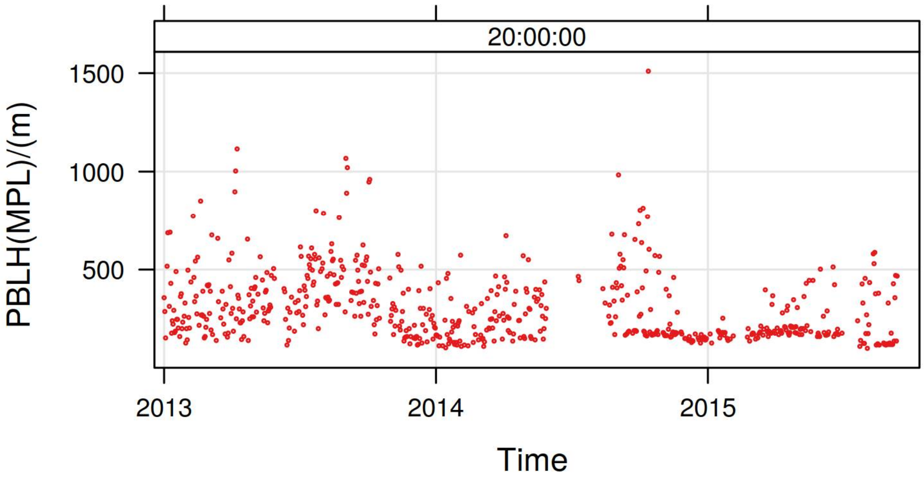

Fig. The retrieved PBL height at 20:00 (LST) from MPL measurements, which presented slight decreasing trend.

Minor Comments

(1) LN33, in->for

LN48-49, I believe these should be in one sentence.

LN153, either remove this sentence or move it to the beginning of this paragraph.

LN329-342, these basic O3 reactions should be put in the introduction, rather than in the results.

LN355-356, or more intensified urbanization in XJH (thus more enhanced downward mixing of O3 (Hu, et al., 2013))

Thanks for the corrections, which have been revised.

(2) anti-correlation for what time period? Nighttime anti-correlation does not indicate VOC-limited mechanism.

The NOx and O_3 measurements were strongly anti-correlated during noontime (10:00-13:00, LST), which has been revised.

(1) LN76, what is the definition of "non-attainment days" in terms of O3 in China? LN81, what is the "Chinese National Ambient Air Quality Stand" in terms of O3?

The "non-attainment days" of O_3 was defined in the ambient air quality standard (GB3095-2012) by the Ministry of Ecology and Environment of the People's Republic of China, providing the condition of daily maximum O_3 concentration exceeding 200 ug/m^3, or daily maximum 8h-O_3 concentration exceeding 100 ug/m^3.

(2) LN104 "However, such O3 variation responding to emission change has not been clearly investigated". You just wrote "Gao et al. (2017) reported that O3

concentration in Shanghai downtown increased 67% from 2006 to 2015, whereas NOx concentration decreased about 38%";

LN120, I thought Gao et al. analyzed 10 yr data as in this study.

Gao et al. (2017) has investigated the O_3 variation during 2006 to 2015 in Shanghai, while only limited in the downtown XJH site. In this study, we calculated and compared the variability of mean daily maximum O_3 concentration, and mean daily maximum 8h-O_3 concentration at downtown site XJH and sub-urban site PD by more comprehensive measurements. In addition, we further illustrated the O_3 increasing trend in the larger scale by using the O_3 measurements from 31 sites over Shanghai, which were not reported by Gao et al. (2017). Furthermore, this study explored the O_3 enhancement response to NOx reduction in Shanghai by WRF-Chem models. The effects of emission changes on long term O_3 variability were evaluated by WRF-Chem and compared with measurements. In addition, the shift of O_3 photochemical regime relative to the variations of NOx and VOCs concentrations in the past ten years was discussed by O_3 isopleths diagram combined with WRF-Chem to provide more insights into the O_3 control strategy. Moreover, the future O_3 levels and its possible chemical regime in Shanghai were also discussed according to the Shanghai Clean Air Action Plan. This was complemented in the part of Introduction.

(3) LN124, high-resolution? Figure 7 looks like very coarse resolution. What is the resolution in Fig. 7?

Fig. 7 should be combined into Fig. 1;

LN141-142, why these point sources do not show up on Fig. 7? Please mark these major point sources in Fig. 7

LN204, Fig. 7 appears to have a resolution coarser than 6km.

The horizontal resolution set in WRF-Chem was 6km. However the emission inventory used in WRF-Chem was extracted and combined from the MEIC (0.25°) and MIRAGE-Shanghai (0.16°) emissions and equally allocated to model grids with 6 km resolution. Thus the emission in Fig.7 seemed to be a little coarse due to the coarse resolution of the emission inventory data. According to the suggestion from the reviewer, the Fig.7 was combined into Fig.1, and the major point source around BS site was marked in Fig.1.

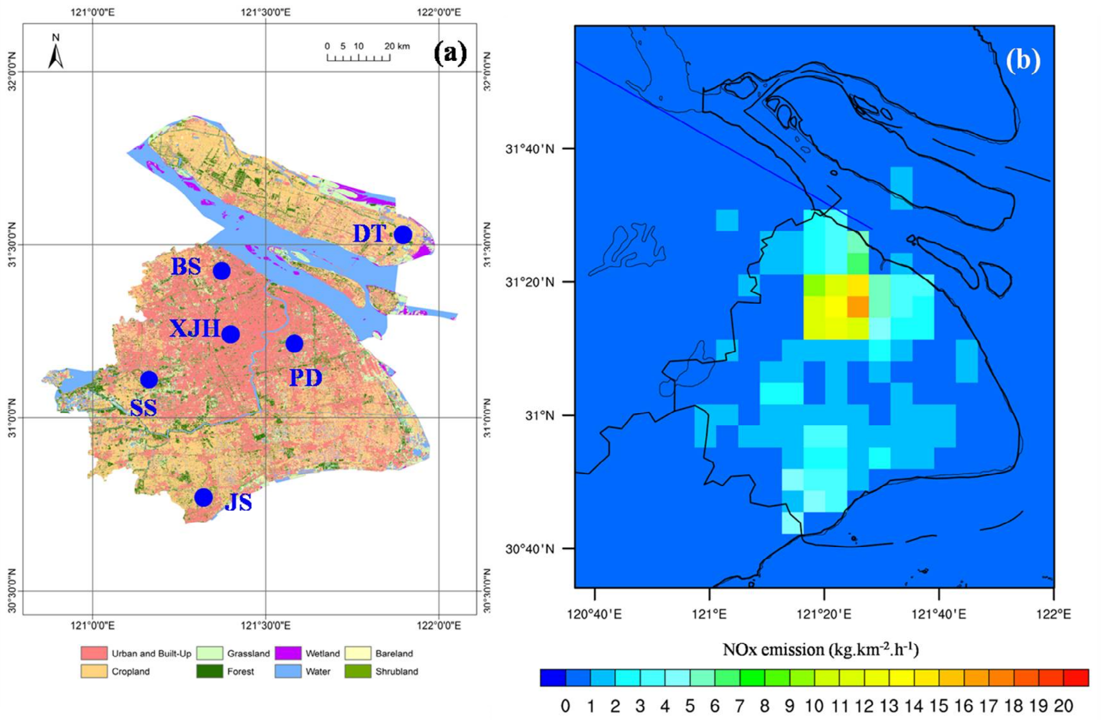

Fig.1 (a) The distribution of land-use category in Shanghai. The blue dots denote the locations of 6 sties (XJH, BS, PD, SS, JS, DT), in which XJH site is located at the downtown of Shanghai, with large emission from transportation, PD site is located at the sub-urban area with the mixed emissions of transportation and residential, JS site is located in the south of Shanghai with several large chemical industries, BS site is located in the north of Shanghai with some big steel and power plants, SS site is located at the top of the sole hill (100 m a.g.l) with influence from regional transport, DT site is located at a remote island without anthropogenic activities. (b) The NOx emission of 2009 scenario in Shanghai.

(4) LN193, what aerosol module?

LN197, what is the “anon-traditional SOA module”

The WRF-Chem model used in this study was not the standard version from the WRF DOWNLOAD website. It was mainly improved by Tie et al. (2007) and Li et al. (2010; 2011). The aerosol module was developed by the US EPA (Binkowski and Roselle, 2003) and used in CMAQ model. The secondary organic aerosol (SOA) formation is simulated using a non-traditional SOA model including the volatility basis-set modeling method in which primary organic components are assumed to be semi-volatile and photochemically reactive and are distributed in logarithmically spaced volatility bins (Li et al., 2011). The partitioning of semi-volatile organic species is calculated using the algorithm suggested by Koo et al. (2003), in which the bulk gas and particle phases are in equilibrium and all condensable organics form a pseudoideal solution (Odum et al., 1996). Nine surrogate species with saturation concentrations from 10^{-2} to 106 μgm^{-3} at room temperature are used for the primary organic aerosol (POA) components following the approach of Shrivastava et al. (2008). The SOA contribution from glyoxal and methylglyoxal is also included (Li et al., 2011). These were added to the revised version.