Response to Referee #2

We thank the anonymous referee for the thoughtful comments and remarks. We provide responses to the referee comments (in bold) below and include the additional references cited at the end of this response:

This work presents aircraft measurements of O3 and its precursors in Hebei Province, China, aiming at understanding the production of ozone within the planetary boundary layer (PBL). They presented vertical profiles of trace gas species, including O3, NOx, CO, and VOCs. A box model was used to relate those concentrations to the O3 production rate and to assess the O3 production and OH reactivity relevant to the VOC/NOx ratio. Their analysis showed that measured O3 levels ranged from 52 to 142 ppbv, with the peak median concentration (~94 ppbv) between 1000 and 1500 m. The NOx concentrations exhibited strong spatial and altitudinal variations, ranging from 0.15 to 49 ppbv. They presented the ratios of CO/NOy and CO/CO2 to indicate the prevalence of low efficiency combustion from biomass burning and residential coal burning. Their measurements of concentrations of total measured VOCs showed that alkanes and alkenes/alkynes were responsible for 74% of the total VOC reactivity, while aromatics contributed the most to the total Ozone Formation Potential (43%) with toluene, m/p- xylene, ethylene, propylene, and i-pentane playing significant roles in the production of O3 in this region. Their box model calculations constrained by measured precursors indicated the peak rate of mean O3 production was ~7 ppbv/hour below 500 m. They also showed that pollution frequently extended above the PBL into the lower free tropo- sphere, where NO2 mixing ratios (~400 pptv) led to net O3 production rates up to ~3 ppbv/hour and this pollution traveled extended distances downwind. They concluded the O3 sensitivity regime as NOx-limited throughout the PBL, while VOC-limited at low altitudes near urban areas. Overall, there are very limited measurements on the vertical profiles of ozone and its precursors as well as an assessment of vertical ozone production and OH reactivity in this region. As such, this work is publishable in ACP, after the following issues have been adequately addressed.

Main points (1) I am surprised that they did not found much contribution from biogenic VOCs between May and June 2016 in this region. Ground-based measurements in NCP have clearly showed a role of BVOCs in ozone and PM production (Wang et al., Use of a mobile laboratory to evaluate changes in on-road air pollutants during the Beijing 2008 summer Olympics, Atmos. Chem. Phys. 9, 8247, 2009; Guo et al., Elucidating severe urban haze formation in China, Proc. Natl. Acad. Sci. USA 111, 17373, 2014). Some comparison with ground-based measurements and discussions of the contribution of BVOCs to ozone production would be essential.

We thank the reviewer for providing the references for the role of BVOCs on ozone and PM production and agree a discussion of BVOCs should be included. We plan to add the following paragraph to the Introduction of the revised manuscript to address this comment, including citation of the studies suggested by the reviewer:

> Natural emissions are the largest source of VOCs globally and react more efficiently with OH than most anthropogenic compounds (Di Carlo et al., 2004), but exhibit a strong

> seasonal, diurnal, and spatial dependence (Li et al., 2013). Biogenic VOCs have been found to play a significant role in the formation of O_3 at the surface (Ma et al., 2019; Zong et al., 2018) and throughout the boundary layer in the NCP (Wang et al., 2008), as well as influence production of $PM_{2.5}$ (Guo et al., 2014) and secondary organic aerosols (SOA) (Wu et al., 2020b). In particular, isoprene has been estimated to account for 27% of the total O_3 production in June 2010 in Beijing (Mo et al., 2018), suggesting the need to consider biogenic isoprene emissions in formulating O_3 control strategies. Quantifying the abundance of NO_x and the suite of VOC chemicals throughout the lower troposphere is urgently needed to better understand the photochemistry of O_3 production in the NCP, which in turn will lead to the development of successful mitigation strategies.

In this new section noted above, we choose not to cite Wang et al. (2009) as the referee suggested since this paper seems to be on a different topic. Instead, we include a publication where Wang et al. (2009) was a coauthor (Mo et al. (2018)), as well as a paper by Q. Wang et al. (2008) in *Science of the Total Environment*, which modeled the impacts of biogenic emissions of VOCs and NO_x on the formation of tropospheric ozone during summertime in eastern China.

In the present study, we were not able to quantify many prevalent biogenic VOCs, such as alpha and beta pinene and monoterpenes; however, we did quantify isoprene. We plan to add a comparison of ARIAs isoprene measurements to the literature in Section 3.2 to the revised manuscript. The new text will read as follows:

> Additionally, our observations have higher amounts of branched alkanes, such as 2,2,4-trimethylpentane and 2-methylheptane (both components of gasoline), but lower amounts of isoprene due to collection over mostly urban regions with lower ambient temperatures than the summer months. Since isoprene with a lifetime of hours (Seinfeld and Pandis, 2006) in the summer typically exhibits a strong vertical gradient in the PBL (Huang et al., 2017), we find the mean amount of isoprene measured during ARIAs is about 7 times lower than average May 2014 surface measurements in Beijing (Li et al., 2015), as well as ~200 pptv lower than June-July 2007 airborne measurements in the PBL in NE China (Xue et al., 2011).

Isoprene has been observed to be important near the surface but since ozone is made throughout the PBL, our observations expand the knowledge base for ozone formation. We add the following new text to Section 3.3.2 of the revised manuscript:

> At a surface site in Beijing (May 2014), Li et al. (2015) found m/p-xylene, ethylene, toluene, propylene, and o-xylene are most influential to OFP, while at a ground station in Tianjin (August 2018), Han et al. (2020) found that ethylene, isoprene, toluene, m/p-xylene, and propylene were important contributors to OFP. Our study supports a larger contribution of anthropogenic VOCs than biogenic VOCs in spring, although summer studies indicate a major role for isoprene to the formation of O_3 in the NCP (Han et al., 2020; Zong et al., 2018). Since isoprene is mostly emitted by biogenic sources during the warmer summer months with strong solar radiation and when soil moisture is sufficient for plant growth, we expect isoprene to have a larger impact on O_3 production in the summer than during spring, the time of our study.

Lastly, we add the following new sentence to the Conclusions:

> In contrast to other surface studies in summer, we find a lower contribution of biogenic sources (e.g. isoprene) to the formation of O_3 in the PBL.

(2) I would also think that the vertical profile in ozone production within the PBL also reflects photochemistry, which is closely related to PBL height and PM levels. Specifically, it has been known that the PBL height strongly regulates the photolysis rate (O1D) and there exists a strong feedback between PBL and PM (An et al., Severe haze in Northern China: A synergy of anthropogenic emissions and atmospheric processes, Proc. Natl. Acad. Sci. USA 116, 8657, 2019; Wu et al., Aerosol–photolysis interaction reduces particulate matter during wintertime haze events, Proc. Natl. Acad. Sci. USA 117, 9755, 2020). Typically, the trends in surface O3 and PM are believed to be anti-correlated. To what extend the PBL-photolysis interaction would impact their assessments of the vertical ozone production and OH reactivity in the present work?

We agree that there are strong feedbacks between the PBL and PM and that the impact of aerosols on ozone production, even the sign of the effect, depends on their optical properties as well as vertical distribution (Dickerson et al., 1997; Kelley et al., 1995). The 0-D box model cannot simulate the depth of the PBL, but since our simulations were constrained by observations, automatically includes effects of dilution due to the height of the PBL. We will add the following revised text to the introduction as well as the recommended references to the manuscript:

> The role of VOCs on the formation of O_3 depends on the characteristics of the environment, including the main emission sources of primary pollutants and ambient temperature (Pusede et al., 2014), and the interaction of aerosols within the PBL to reduce photolysis (An et al., 2019). High aerosol concentrations have been shown to decrease photolysis and hinder summer surface O_3 formation by 25 ppbv on average in Xi'an, China (Feng et al., 2016), which pose a challenge for pollution control strategies.

The net impact of j(O3) and j(NO2) and thus the rate of ozone production was tested for in our model calculation, but is small. We also add new text to Section 2.2 describing the impact of aerosols on vertical ozone production:

> The impact of aerosols on O_3 production depends on the optical properties as well as the vertical distribution (Dickerson et al., 1997; Kelley et al., 1995). In the presence of scattering and absorbing aerosols, photolysis frequencies will be altered, thus changing the O_3 formation and atmospheric oxidizing capability (Wu et al., 2020a). Previous research over China has shown that as AOD increases, the extinction effect of aerosols on photolysis frequencies decreases due to a higher proportion of scattering aerosols under high AOD conditions (Wang et al., 2019a). Optical depth, single scattering albedo, and angstrom exponent during ARIAs (see Wang et al., 2018a) are used in the TUV online calculator (https://cprm.acom.ucar.edu/Models/TUV/Interactive_TUV/) to assess the impact of aerosols on photolysis frequencies. Most of the aerosol particles during ARIAs were concentrated in the lowest 2 km of the atmosphere with a single scattering albedo at

550 nm of 0.85 and an average AOD ~0.2. The impact of aerosol optical properties measured during ARIAs on photolysis frequencies is small compared to the default setting, so no additional adjustments are made to the model values.

The OH reactivity calculation in this manuscript uses rate constants published by MCM and NIST, which represent optimal conditions in which there are no aerosols. We will add the following text to Section 3.3:

> In this section, we present results using the loss rate of each VOC species with OH and ozone formation potential (OFP) assuming no influence of aerosols. Since the aerosol effect on O_3 formation is dependent upon time of day (solar zenith angle), meteorology, levels of local and neighboring aerosols, and the VOC/NOx ratio, the calculations presented here are simplified compared to the more complicated chemical composition of the atmosphere, but are still useful to help inform control strategies.

We will add the following sentence to the conclusions to stress the importance of better understanding the aerosol impact of ozone production:

> The photochemistry of O_3 production is highly dependent upon the interaction of radiation and aerosols within the PBL and future work is needed to assess optical properties of aerosols at wavelengths relevant to photolysis of O_3 to $O(^1D)$ and thus OH.

(3) Their measurements were made between May and June 2016. Recent studies have shown significantly different trends in O3 and PM precursors (particularly in NOx) in this region (Zhang et al., An unexpected catalyst dominates formation and radiative forcing of regional haze, Proc. Natl. Acad. Sci. USA 117, 3960, 2020). How did their measurements fit into those of trends for O3 and PM precursors?

We agree with the referee that several recent studies have found different trends in O3 and PM precursors in this region and have added the following discussion at the end of the Introduction:

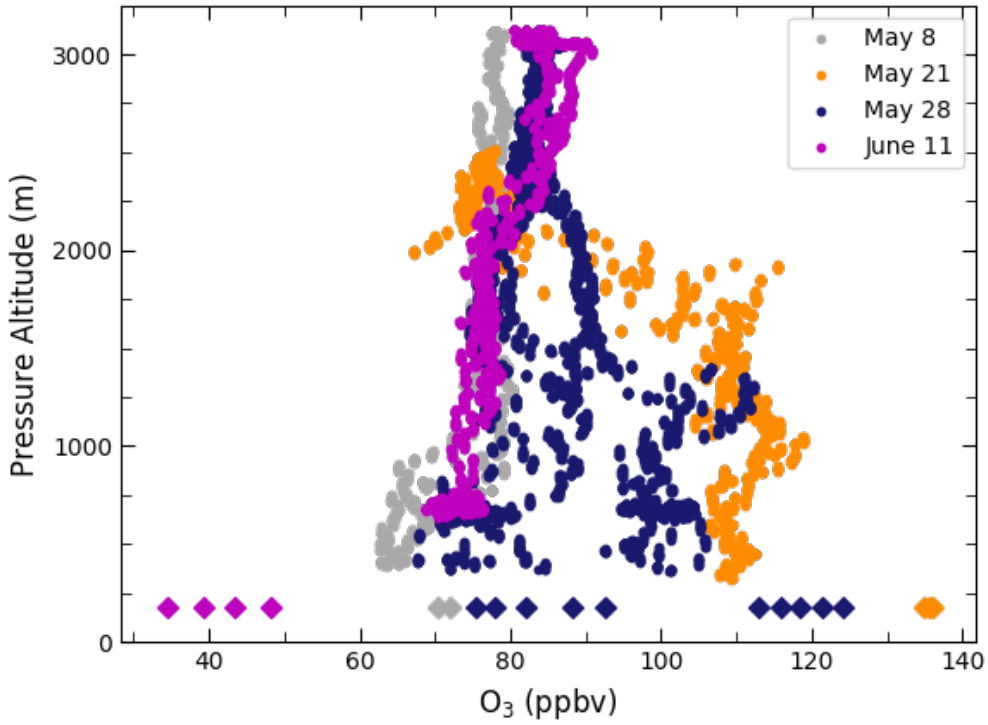
> The North China Plain is one of the most polluted regions in the world, but implementation of pollution reduction measures through the Five-Year Plans has allowed for decreasing trends of many pollutants. In particular, Zhang et al. (2020) found an increased number of days of clean/light haze and a decreased number of days with heavy haze, along with a significant decline of SO_2 concentrations. Similarly, using observations from MODIS and OMI, Si et al. (2019) found AOD and SO_2 to decrease from 2006 to 2015, while NO_2 rose by 4.79% in the NCP during this period. While surface NO_2 decreased 20% from May 2014 to December 2018 throughout China, there are still a large number of measurement stations with increasing trends of NO_2 due to changes in meteorological conditions and aerosol emissions (Fan et al., 2020), illustrating the need for more research characterizing air pollution in this region.

Since our measurements are limited to a small part of Hebei Province from 11 research flights in May and June 2016, we compare our aloft observations to surface ozone concentrations from the

A^2BC site located in Xingtai. We will add the following new text to Section 3.1 of the revised manuscript with the following figure, which will be *Figure S3,* included in the supplement:

> The vertical profiles of O_3 compared to concurrent surface measurements in Xingtai indicates the A^2BC site usually observed larger average concentrations than observed aloft, but this difference was highly dependent upon time of day (Fig. S3). The early afternoon profiles on May 8 showed average surface concentrations only slightly higher than the Y-12 measurements at ~400 m, while the mid-afternoon profiles on May 21 showed ~25 ppbv higher surface O_3 concentrations than Y-12 observations. At low altitudes (~700 m), the late morning flight (around 11:00 LST) on May 28 observed levels of O_3 ranging from 72-80 ppbv, comparable to average surface concentrations of 78 ppbv at the same time. By contrast, the afternoon flight (approximately 17:00 LST) at the same altitude later that day observed ~25 ppbv lower levels of O_3 compared to the surface (average=121 ppbv). All profiles on June 11 showed 10-30 ppbv lower average surface concentrations than measured during the Y-12 spirals.

Figure S3. Vertical profiles (N=20) of 1-second O_3 concentrations (ppbv) from the Y-12 (circles) compared to concurrent average concentrations measured at the A^2BC site in Xingtai (diamonds). The average surface O_3 concentration was computed by averaging the 5-minute data interval starting 30 minutes before the spiral until 30 minutes after the spiral was completed.



Minor points: In general, the paper was reasonably-well written, but could be further improved to increase its readability. Below are a few examples. (1) The usage between past and presented tenses was interchangeable, but should be made consistent throughout the manuscript. "This analysis shows measured O3 levels ranged from ... The NOx concentrations exhibited ... Ratios of CO/NOy and CO/CO2 indicate . . ."

We thank the reviewer for these readability suggestions. The present tense was used for the presentation of the analysis, while the past tense was used when discussing the airborne observations. The idea is to distinguish measurements (past tense) from general conclusions (present tense), i.e., concentrations were X implications are Y. We have decided to keep this style in the manuscript.

(2) The phrase "26 whole air canisters" in the abstract was confusing.

We will remove the phrase "26 whole air canisters" from the Abstract.

(3) The phrase "we see evidence of" in the abstract was rather causal.

We will revise this sentence to remove the casual phrase in the Abstract to now read:

> Ratios of CO/CO_2 indicate the prevalence of low efficiency combustion from biomass burning and residential coal burning, but indicate some success of regional pollution controls compared to earlier studies in China.

(4) The sentence "demonstrating both VOCs and NOx need further control to reduce aloft O3" in the abstract needs to be re-written.

We will change this phrase to read "demonstrating that control of both VOCs and NOx is needed to reduce aloft O_3 pollution over Hebei."

Lastly, we also became aware of a recently published paper on air quality in China by Souri et al. (ACP, 2020). This paper focused on satellite observations and is an ideal complement to our airborne based analysis. Upon revision we propose to cite this paper in the following manner in Section 3.4:

> Using updated emissions from a nonlinear joint analytical inversion of VOCs and NO_x from the Ozone Mapping and Profile Suite Nadir Mapper (OMPS-NM) formaldehyde and OMI NO2 columns during KORUS-AQ in WRF-CMAQ, Souri et al. (2020) found the maximum daily 8 hour average surface O_3 over the NCP to increase by 4.56 ppbv, suggesting that emission control strategies on VOCs should be prioritized.

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