Response to Reviewer 1:

This paper presents a detailed analysis of the HONO budget in the Pearl River Delta region of China. The paper is well written, the data and the analysis are well presented. The subject is fit for publication in ACP and I would recommend the paper is accepted after the authors have addressed the following concerns.

Author's Response: We would like to thank the reviewer for the helpful comments and suggestions. We have carefully considered all the comments and revised the manuscript accordingly. For clarity, we list the original reviewer's comments below *in black italic*, and provide our responses and changes in the manuscript in blue and red, respectively.

Major comments:

1. Please add more information about the box-model. The MCM is not a model, it is just the chemical mechanism used in a model. Which software/modelling tool was used? Which VOCs were included? How was photolysis calculated for the non-measured photolysis rates? Were other processes (heterogeneous, deposition, etc..) included?

Author's Response: We thank the reviewer for the constructive comments. Accordingly, more detailed information about the box-model is introduced as follows:

To evaluate the influence of HONO chemistry on the atmospheric oxidation capacity, a zero-dimensional photochemical box model (Framework for 0-Dimensional Atmospheric Modeling–F0AM) based on the Master Chemical Mechanism (MCMv3.3.1) (Wolfe et al., 2016; Jenkin et al., 2003; Jenkin

et al., 2015) was applied to calculate the concentrations of O_3 and OH radicals. The model was implemented in MATLAB 2012. The hourly averaged measurement data were used to constrain the simulation, including HONO, NO, NO_2, CO, SO_2, VOC species (listed in Table S1), temperature, water vapor, wind speed, wind direction, pressure and photolysis frequencies $J(NO_2)$, $J(HONO)$, $J(O^1D)$ and $J(H_2O_2)$. Other non-measured photolysis frequencies were calculated according to Eq. (1) (Jenkin et al., 1997), and then scaled by the measured $J(NO_2)$:

$$J_i = L_i \cos(\chi)^{M_i} exp\left(-N_i sec(\chi)\right) \tag{1}$$

where χ represents the solar zenith angle (SZA); L_i, M_i and N_i are the photolysis parameters under clear sky conditions which were taken from Jenkin et al. (1997). The heterogeneous processes as well as deposition of chemical species were not considered in this model.

Table S2. The VOCs species constrained in the F0AM model.

Classification	Measured hydrocarbons
Alkane	CYCLOHEXANE, ETHANE, N-BUTANE, N-DECANE, N-NONANE, N-OCTANE, PROPANE, 2-METHYLHEXANE, 2-METHYLPENTANE, 3-METHYLHEXANE, 3-METHYLPENTANE, 2-METHYLPROPANE, 2-METHYLBUTANE, PENTANE, HEXANE, HEPTANE, HENDECANE
Alkene	PROPENE, TRANS-2-BUTENE, TRANS-2-PENTENE, 1-BUTENE, 1-PENTENE, 1-HEXENE, CIS-2-BUTENE, CIS-2-PENTENE, STYRENE
ISO	ISOPRENE
Alkyne	ETHYNE
Aromatic	BENZENE, N-PROPYLBENZENE, 1-2-3-TRIMETHYLBENZENE, 1-2-4-TRIMETHYLBENZENE, 1-3-5-TRIMETHYLBENZENE, METHYLBENZENE, ETHYLBENZENE, 1,4-DIMETHYLBENZENE, 1,2-DIMETHYLBENZENE, I-PROPYLBENZENE, 1-ETHYL-3-METHYLBENZENE, 1-ETHYL-4-METHYLBENZENE, 1-ETHYL-2-METHYLBENZENE

2. The molybdenum converter used to measure NOx is subject to known interferences by other NOy species. Since a large part of the analysis in this paper relies heavily on NO and NO2 data, this issue cannot be neglected. I would expect the interference to be significant under the urban conditions considered here. The authors should address this issue and examine how the results of the studies are affected by it.

Author's Response: Thanks for the suggestion. We are aware that the chemiluminescence technique combined with molybdenum converter, albeit widely used to detect NO and NO_2, suffers from the interference of some reactive nitrogen species (NOy) like HNO_3, HONO, Peroxyacetyl nitrate (PAN), other Organic nitrate, N_2O_5, etc., which can be reduced to NO by the molybdenum converter, leading to an overestimate of NO_2 concentration. The degree of overestimation depends on both air mass age and the composition of the oxidation products/intermediates of NOx. At urban sites that are greatly affected by fresh emissions, such interference has been estimated to be 3%−10% (Xu et al., 2013; Dunlea et al., 2007; Villena et al., 2012), while it could be substantially higher at the suburban sites that receive aged pollution (~30–50%) (Xu et al., 2013), even up to 100% or more at some sites with the mostly aged pollution air (Dunlea et al., 2007; Steinbacher et al., 2007).

Our site is a typical urban site with heavy traffic emissions, as indicated by high concentrations of NO and NOx. Meanwhile, the average concentration of HONO, gaseous HNO_3 and particulate nitrate during the campaign were 0.74 ± 0.70 ppbv, 2.1 ± 2.0 ppbv and 4.2 ± 5.8 μg m^{-3}, respectively. PAN was not measured and is estimated around 0.84 ppbv based on earlier data at Guangzhou (Wang et al., 2015) and the other NOy species can be ignored. Based on these, we roughly estimate the relative interferences of NOz (NOy-NOx) to NO_2 to be around 10%. We believe such a discrepancy would not affect the validity of our findings, and the following statements have been added in our revised manuscript to acknowledge the limitation of the measurement technique.

It should be noted that the molybdenum oxide (MoO) converters may also convert some NOz (= NOy - NOx) (e.g., HONO, peroxyacetyl nitrate (PAN), HNO_3, and so on.) species to NO and hence could overestimate the ambient NO_2 concentrations. The degree of overestimation depends on both air mass

age and the composition of NOy. At our site that was greatly affected by fresh emissions, the relative interferences of NOz to NO_2 have been estimated to be around 10%, which is closed to the results of Xu et al. (2013) and negligible for our discussion of HONO budget.

3. I think the discussion in section 3.2 needs to be improved. First the observed HONO production rate should be presented and shown (how was it calculated, which are the mean values, etc..). This will make the following calculations easier to understand. Besides that, I have two main comments regarding this section.

One, the authors infer that a large missing sink of HONO is required to explain the observations (lines 274-275). However, their calculation of HONO primary emissions relies on emission inventories that are likely not very accurate. The possibility that HONO primary emissions are overestimated in the emission inventories cannot be neglected and needs to be discussed.

Two, the authors are deriving a primary emission rate of 0.04 ppb/h or more (line 272), a soil emission rare of 0.02 ppb/h (line 297) and a net production via OH+NO of 0.26 ppb/h (line 314), while the average observed HONO production rate is 0.02 ppb/h (line 271). From this an unknown sink of 0.25 ppb/h is inferred. First of all, in order to close the budget, the unknown sink should be 0.30 ppb/h (unless you mean that 0.05 ppb/h is lost via deposition, it is not clear from section 3.2.4). More importantly, the discussion in section 3.2.3 implies an additional, non quantified source due to NO2 reaction on surfaces, so the unknown sink is actually a lower limit (but see also the previous comment, regarding possible overestimation of primary emissions). These calculations should be make clearer, maybe with an extra "summary" subsection at the end of section 3.2.

Author's Response: Thanks for the suggestions. A number of changes have been made accordingly as follows.

First, as suggested by the reviewer, we have introduced the observed HONO production/accumulation rate in our revised manuscript:

The observed HONO production/accumulation rate P_{HONO} is calculated by Eq. (4):

$$P_{HONO} = \frac{[HONO]_{t2} - [HONO]_{t1}}{t_2 - t_1} \tag{4}$$

where $[HONO]_{t1}$ and $[HONO]_{t2}$ represent the HONO concentration at 18:00 and 6:00 Local Time, respectively. Then an average P_{HONO} of 0.02 ppbv h^{-1} can be derived.

In response to the other two comments:

(1) The uncertainty of the two sets of inventories is in the range of -25%–28% (Huang et al., 2021), leading to an uncertainty of HONO primary emissions. The HONO emission rates obtained at nighttime are 0.3 ± 0.15 ppbv h^{-1} and 0.04 ± 0.02 ppbv h^{-1}, as the upper and lower limits of the nighttime HONO emission rate, respectively, which are both larger than the observed HONO accumulation rate (0.02 ppbv h^{-1}). On the other hand, P_{emis} only contributed a minor fraction (8%) to the daytime HONO sources. Therefore, even though the HONO primary emissions are overestimated in the emission inventories, the results of HONO budget would not change much. As the reviewer suggested, the uncertainty of the inventories and its possible impact have been discussed in the revised manuscript as follows:

Considering the uncertainty of the inventories (-25%–28%), P_{emis} may be overestimated or underestimated to the same extent. Nevertheless, direct emission of HONO is still a large HONO source at night along with other sources of HONO that remain to be considered.

(2) We thank the reviewer for point out this error. Indeed, as the reviewer stated, a sink of 0.30 ppbv h^{-1} is required to close the budget, and it's just a lower limit. We re-checked the calculation process and found another error in deposition velocity, and the new value of V_d is 1.8 cm s^{-1}. We have revised the manuscript, and added an extra summary for section 3.2:

(2) except for HONO + OH, the strength of HONO sink should be at least 0.30 ppbv h^{-1}, 6 times larger than that obtained by Li et al. (2012) and comparable to that by Hao et al. (2020).

In sum, primary emission from vehicle exhaust (between 0.04 ± 0.02 ppbv h^{-1} and 0.30 ± 0.15 ppbv h^{-1}) and the homogeneous reaction of OH + NO (0.26 ± 0.08 ppbv h^{-1}) were major sources of HONO at night. Nighttime soil emission rate was calculated to be 0.019 ± 0.001 ppbv h^{-1}, which is comparable to the observed nocturnal increase rate of HONO (0.02 ppbv h^{-1}), further indicating the importance of direct emissions. Additionally, contribution from NO$_2$ heterogeneous reactions on surfaces should not

be ruled out. To balance the nighttime HONO budget by assuming dry deposition to be the principal loss process, a dry deposition rate of at least 1.8 cm s^{-1} is required.

4. In section 3.4, I would suggest that if VOC data are available, than ozonolysis of alkenes should be added here. Several studies have suggested that these process may be important in urban conditions. In fact, why not use the model results from section 3.5 to calculate the OH production pathways? It would be more comprehensive than what is shown in figure 9.

Author's Response: Thanks for the suggestion. We have added the results of the reaction of ozonolysis of alkenes in Figure 9. Compared with HONO and O$_3$, this pathway seems insignificant at our site. Table S5 summarized the reaction rate constant of O$_3$ with alkenes at 298 K and the yields of OH. Results from model would be more comprehensive but suffer from uncertainties of other processes. Calculating the OH radicals' production rate based on observational data is a direct and standard method that was applied in many studies (Zheng et al., 2020; Shi et al., 2020; Su et al., 2008; Liu et al., 2019; Yang et al., 2014; Ren et al., 2020; Alicke et al., 2002; Ge et al., 2021; Heard et al., 2004). Therefore, we also adopted this method and made a comparation with similar researches.

$$P_{(O_3+alkenes)-OH} = \sum k_{alkenes(i)+O_3}[alkenes(i)][O_3]\,Y_{OH_i} \tag{13}$$

In Eq. (13), $k_{alkenes(i)+O_3}$ represents the reaction rate constant for the reaction of O$_3$ with alkene (i), Y_{OH_i} represents the yield of OH from the gas-phase reaction of O$_3$ and alkene (i). Table S5 summarized the reaction rate constant of O$_3$ with alkenes at 298 K and the yields of OH. In daytime, the sum of OH production rates by ozonolysis of alkenes was 3×10^5 cm^{-3} s^{-1}, which is much smaller than that of HONO and O$_3$. This value (3×10^5 cm^{-3} s^{-1}) was comparable to the results in previous studies (Kim et al., 2014; Ge et al., 2021; Martinez et al., 2003; Ren et al., 2003; Lee et al., 2016; Alicke et al., 2002; Kleffmann et al., 2005; Ren et al., 2013), but smaller than some other studies (Shi et al., 2020; Zheng et al., 2020; Heard et al., 2004).
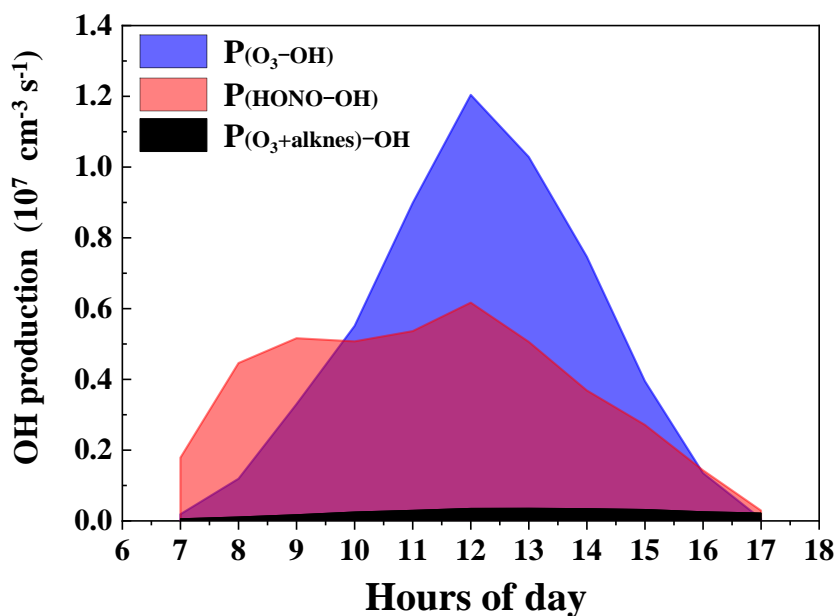
Figure 9. The yield and comparison of OH radicals by HONO, O$_3$ and ozonolysis of alkenes.

Table S5. Ozonolysis reaction rate constants and OH formation yields of the volatile organic compounds (VOC) used in the calculation.

VOC	k (298 K)/($\times 10^{-18}$ cm^3 molec.$^{-1}$ s^{-1})[a]	OH yield
PROPENE	10.1	0.34[b]
TRANS-2-BUTENE	190	0.59[b]
TRANS-2-PENTENE	160	0.47[c]
1-BUTENE	9.64	0.41[b]
1-HEXENE	11.3	0.32[b]
1-PENTENE	10.6	0.37[b]
CIS-2-BUTENE	125	0.37[b]
CIS-2-PENTENE	130	0.3[c]
STYRENE	17	0.07[c]
ISOPRENE	12.8[c]	0.13 ± 0.03[c]

[a] Atkinson and Arey (2003); [b] Rickard et al. (1999); [c] Alicke et al. (2002)

Minor comments:

1. lines 169-171: what does it mean that "the boundary layer diurnal cycle has been modified"? And what are the "solar altitude" and the "photolysis rate correction coefficient"?

Author's Response: The boundary layer diurnal cycle is a common application of the F0AM (Wolfe et al., 2016). It reflects the meteorology at the site where the model is applied for, and was modified to represent local geographical and meteorological conditions at our site.

"Solar altitude" in the original manuscript is inaccurate, and has been replaced by "solar zenith angle (SZA)". Photolysis rate correction coefficient is the ratio between measured $J(NO_2)$ and calculated $J(NO_2)$.

2. figure 3: a blue line with pink shading is confusing. It would be better to use a shade of blue. Also why not add the results obtained with the other two methods? It may be interesting to compare them.

Author's Response: Thanks for the suggestions. We have changed the color of the shade of the blue line in Figure 3 to light blue (see below). Basically, the results obtained by method (1) is not the HONO primary emission rate, but assumed HONO concentrations caused by emissions, which are often compared with observed HONO concentration to get the relative contribution of emissions. Additionally, we think that the calculation of method (2) is problematical, because a negative primary emission rate would be derived in NOx decline period, which is unrealistic. The result obtained by method (3) is the rate of change in HONO concentration, which is in line with the definition of emission rate. Specially, the unit of the quantity obtained from method (1) is ppbv, while that from method (3) is ppbv h^{-1}. They are not comparable.
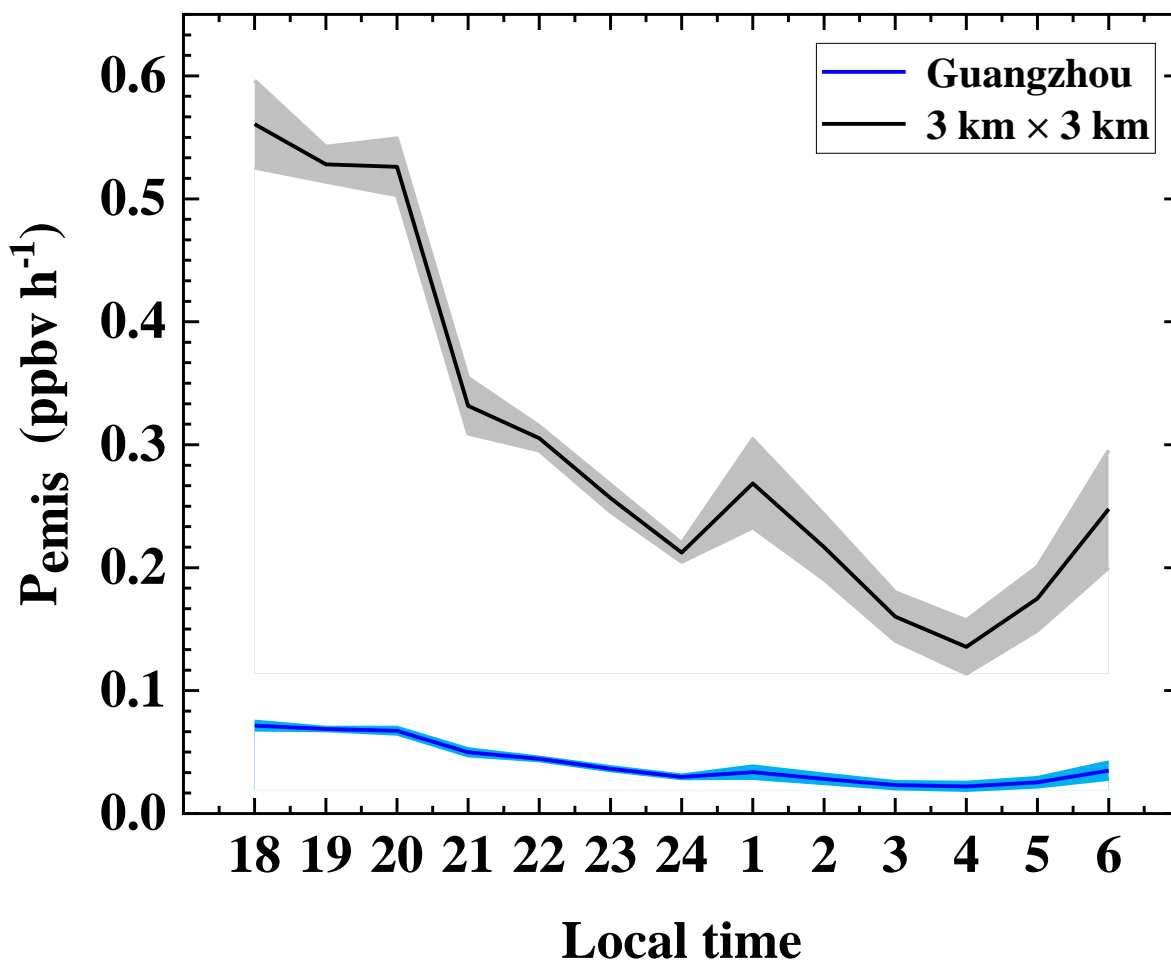
Figure 3. The nocturnal variation of HONO primary emission rates. The black and blue lines represent the HONO primary emission rates calculated by the 2017 NOx emission source inventory of the 3 km × 3 km grid cell centred on the Guangzhou Institute of Geochemistry and the 2017 NOx emission source inventory of Guangzhou city respectively. The coloured areas represent 1 – σ standard deviations.

3. figure 5: I would not consider the correlation netween HONO and NO, "a good correlation". In fact

it is not even linear, meaning it doesn't really provide evidence that OH+NO is a major pathway.

Author's Response: We agree that the relationship between HONO and NO is non-significant linear. We found that under conditions with low NO concentration, high concentration of HONO also appeared, suggesting processes other than primary emissions or homogeneous reaction of OH + NO were involved in HONO formation. Nevertheless, the correlation ($R^2 = 0.5927$) is relatively high compared to many nighttime observations (Zhang et al., 2019; Fu et al., 2019; Yang et al., 2014; Yang et al., 2021; Tong et al., 2016; Ge et al., 2021; Li et al., 2021). Although correlation cannot indicate the causal relationship between HONO and NO, it does suggest that these two may be related. The original text has been revised to convey this point more accurately.

The relatively high correlation ($R^2 = 0.5927$) between HONO and NO is in line with this finding (Fig. 5 (a)).

4. figure 6: can you explain why you are averaging only the top five HONO/NO2 values?

Author's Response: In the study of Stutz et al. (2004), the pseudo steady state (PSS) of $HONO/NO_2$, which is characterized by a maximum $HONO/NO_2$ ratio, was interpreted as the balance between the heterogeneous conversion of NO_2 to HONO and the loss of HONO on surfaces. For each RH, $HONO/NO_2$ ratios can be influenced by the time of the night, the surface to volume ratio, and other parameters, such as advection. Concentrating solely on these maxima (reaching pseudo steady state) will therefore eliminate much of the uncertainty connected with the influence of other parameters (like the time of the night, the surface density, advection, etc.).

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