Anonymous Referee #2

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This manuscript describes constrained photochemical modeling of four large urban areas in China. The paper is difficult to read due to organization, presentation and grammar. I found it difficult to understand what exactly was modeled or measured and how. The conclusion that all four cities are VOC limited is probably correct and probably worth noting for these cities. However, some of the conclusions such as the importance of some radical sources is more difficult to justify as they are not based on observations. I have included major and minor comments below. However, please note the manuscript needs significant editing for style and grammar beyond my suggestions. I think this paper is only publishable after major revisions.

Answer:

We thank the comments and suggestions from the reviewers, which help to improve the manuscript considerably. The response and changes are listed below. We also changed the style and grammar of the paper and please find them in the revised manuscript.

1) The title and abstract indicate that the "atmospheric oxidation capacity" is the focus of the paper. That is fine but this term should be defined instead of vaguely described as in the first line of the abstract. Once defined the values for the different cities should be reported – preferably in the abstract and in the results. I would define the AOC as the reactions of OH, ozone, NO3, etc. that lead to oxidation of an atmospheric component. I would expect units of something like the amount of oxidized molecules per time. The authors only include OH in their reporting of AOC and only vaguely report the values. This needs to be tightened up. I am sure that OH dominates but ozone and NO3 may be important at night and this should needs to be at least mentioned.

Answer:

In the revised manuscript, we restrict the oxidation capacity to daytime photochemical reaction. Also, we point out the focus of this study will be mainly on OH radical chemistry. We changed the title to be "Daytime atmospheric oxidation capacity in four Chinese megacities during photochemical polluted season: a case study based on box model simulation".

In the beginning of the abstract, we changed the Line 23 Page 1 “Atmospheric oxidation capacity AOC is the core of converting freshly-emitted substances to secondary products, which are dominated by reaction with hydroxyl radicals (OH) during daytime.” We added a sentence to define AOC and restrict it to OH oxidation only in Line 35 Page 2 “AOC can be defined as the sum of respective oxidation rates of trace gases (VOCs, CO, and NO$_x$) by the oxidants (OH, O$_3$, and NO$_3$) (Geyer et al., 2001). Given the relatively importance of OH oxidation during daytime, the AOC is restricted to OH oxidation in this study.”

2) When I read the abstract, I thought this was going to be more of an observational study than a modeling project. I expected to see observations of OH, HO2, etc. So I recommend stating clearly that this is a photochemical modeling study constrained by observations of NOx, ozone, etc. For example, I initially thought that OH reactivity was measured in this study instead of being calculated from VOC observations. So please make it clear what is measured and how. The lack of any detail in the instrumentation section is unacceptable in my opinion. I suggest that a table be made of every parameter that is measured, including the method, and a reference. I realize standard commercial instruments perform some of the measurements such as ozone and CO. However, many of the measurements are not run of the mill. In particular, there needs to be a reference to the VOC measurement method and a list of measured compounds and detection limits listed in the supporting information.

Answer:

As mention previously, we changed the title to be more explicitly demonstrating this is a model-based study. We added a table to describe the instrumentation in the supplement (Table S1). We added a sentence in Line 26 Page 3 “The performance of different instruments is summarized in Table S1.”

Table S1 measured species for ozone pollution analysis and instrument time resolution, accuracy and limit of detection

Species	Method	Time resolution	Accuracy (1σ)	Limit of Detection / ppbv

Photolysis frequencies	Actinic flux spectroradiometry	20 s	±10 %	Five orders of magnitude lower than maximum at noon
O_3	UV absorption	1 min	5%	0.5
NO	Chemiluminescence	1 min	±20 %	60 pptv
NO_2	Chemiluminescence	1 min	±20 %	0.3
CO	IR absorption	1 min	5%	4
VOCs	Gas chromatography and mass spectroscopy /flame ionization detector	1 h	10%~20%	0.01~0.2

We also prepared a table in supplement to state what are measured, modelled, and parameterized in this study (Table S2). The measured VOCs and their concentrations are presented in table S3. We added a sentence in Line 34 Page 3 “The measured, modelled and parameterized parameters are summarized in Table S3.”

We added a description about the VOC measurement in Line 26 Page 6 “VOC measurements (including 55 organic species) were performed by commercial an instrumentation using gas chromatograph (GC) equipped with a mass spectrometer (MS) and a flame ionization detector (FID). In principle, the air sample was drawn into two parallel channels for enrichment by cooling before analysis (Wang et al., 2014). The VOCs measurements include C_2–C_{11} alkanes, C_2–C_6 alkenes, and C_6–C_{10} Aromatics (Table S2).”

Table S2 Summary of measured VOCs concentration for four campaigns

	Beijing			Shanghai			Guangzhou			Chongqing		
VOC / ppbv	**Mean**	**Median**	**Max**	**Mean**	**Median**	**Max**	**Mean**	**Median**	**Max**	**Mean**	**Median**	**Max**
1,2,3-TRIMETHYLBENZENE	0.026	0.022	0.100	0.130	0.120	0.340	0.090	0.065	0.473	0.068	0.053	0.189
1,2,4-TRIMETHYLBENZENE	0.098	0.085	0.370	0.160	0.150	0.610	0.199	0.121	1.140	0.225	0.161	0.756
1,3,5-TRIMETHYLBENZENE	0.022	0.018	0.111	0.004	0.000	0.230	0.077	0.053	0.347	0.079	0.058	0.337
1-BUTENE	0.167	0.140	0.803	0.072	0.060	0.300	0.239	0.218	0.607	0.193	0.146	0.939
1-HEXENE	Nan	Nan	Nan	0.323	0.280	1.870	0.074	0.048	0.429	0.065	0.062	0.193
1-PENTENE	0.025	0.019	0.156	0.049	0.020	0.330	0.049	0.033	0.295	0.057	0.042	0.471
2,2,4-TRIMETHYLPENTANE	0.051	0.045	0.240	0.155	0.130	0.990	0.072	0.039	0.736	0.056	0.045	0.177

2,2-DIMETHYLBUTANE	0.020	0.019	0.070	0.149	0.140	0.410	0.099	0.063	0.898	0.054	0.037	1.236
2,3,4-TRIMETHYLPENTANE	0.021	0.019	0.094	0.023	0.000	0.350	0.045	0.028	0.336	0.026	0.022	0.071
2,3-DIMETHYLBUTANE	0.033	0.028	0.137	0.071	0.070	0.180	0.137	0.072	1.584	0.077	0.058	0.769
2,3-DIMETHYLPENTANE	0.049	0.038	0.469	0.027	0.000	0.490	0.111	0.061	0.667	0.056	0.040	0.353
2,4-DIMETHYLPENTANE	0.039	0.037	0.099	0.112	0.100	0.350	0.070	0.046	0.379	0.030	0.024	0.142
2-METHYLHEPTANE	0.016	0.014	0.050	0.002	0.000	0.210	0.066	0.046	0.440	0.039	0.032	0.162
2-METHYLHEXANE	0.061	0.055	0.227	0.000	0.000	0.000	0.273	0.174	1.391	0.133	0.095	0.976
2-METHYLPENTANE	0.226	0.206	0.983	0.265	0.230	1.610	1.066	0.557	8.730	0.360	0.268	3.827
3-METHYLHEPTANE	0.021	0.019	0.066	0.095	0.100	0.210	0.054	0.037	0.345	0.024	0.020	0.113
3-METHYLHEXANE	0.107	0.093	0.307	0.116	0.110	0.260	0.299	0.177	1.936	0.150	0.101	1.196
3-METHYLPENTANE	0.277	0.252	1.027	0.130	0.110	0.580	0.716	0.378	4.242	0.363	0.259	4.246
BENZENE	0.909	0.780	7.830	0.413	0.350	1.240	0.989	0.560	11.448	1.080	0.995	3.749
CIS-2-PENTENE	0.005	0.004	0.045	0.015	0.000	0.670	0.014	0.007	0.107	0.023	0.005	0.287
CIS-BUTENE	0.035	0.019	0.301	0.003	0.000	0.280	0.122	0.124	0.259	0.143	0.100	1.333
CYCLOHEXANE	0.079	0.058	1.048	0.097	0.080	0.320	0.222	0.103	2.180	0.079	0.064	0.293
CYCLOPENTANE	0.125	0.117	0.355	0.048	0.050	0.150	0.117	0.108	0.313	0.167	0.135	0.716
ETHANE	4.896	4.570	13.941	2.432	2.300	7.570	1.952	1.661	5.029	5.145	4.957	11.305
ETHENE	2.210	2.087	7.887	0.921	0.700	5.290	1.522	1.242	6.875	4.039	3.435	11.949
ETHYLBENZENE	0.335	0.257	1.636	0.355	0.290	1.460	1.322	0.782	16.959	0.625	0.480	2.176
ETHYNE	Nan	Nan	Nan	0.025	0.020	0.130	1.355	1.263	2.949	4.123	3.649	11.352
ISO-BUTANE	1.836	1.747	6.574	0.779	0.650	3.760	1.884	1.536	6.630	0.652	0.542	3.581
ISO-PENTANE	1.414	1.326	3.941	0.691	0.560	3.110	1.205	1.079	5.581	1.987	1.412	34.131
ISO-PROPYLBENZENE	0.011	0.010	0.056	0.033	0.000	0.940	0.047	0.037	0.230	0.032	0.026	0.096
ISOPRENE	0.272	0.208	1.289	0.000	0.000	0.110	0.126	0.088	0.809	0.404	0.332	1.641
M-DIETHYLBENZENE	Nan	Nan	Nan	0.217	0.190	0.820	0.036	0.035	0.181	0.026	0.020	0.088
M-ETHYLTOLUENE	0.052	0.045	0.206	0.033	0.000	0.500	0.168	0.122	0.779	0.150	0.111	0.666
M,P-XYLENE	0.604	0.413	3.006	0.565	0.420	3.180	1.508	0.770	24.621	0.655	0.511	2.352
METHYLCYCLOHEXANE	0.074	0.056	0.344	0.003	0.000	0.460	0.187	0.085	2.100	0.189	0.064	4.341
METHYLCYCLOPENTANE	0.121	0.107	0.399	0.064	0.050	0.190	0.296	0.161	2.022	0.120	0.088	0.776
N-BUTANE	2.579	2.403	8.366	0.770	0.600	3.360	2.790	2.339	9.093	1.050	0.847	6.242
N-DECANE	0.021	0.018	0.093	0.074	0.070	0.270	0.108	0.071	0.544	0.086	0.068	0.206
N-HEPTANE	0.116	0.095	0.386	0.037	0.000	0.310	0.197	0.113	1.420	0.209	0.158	1.230
N-HEXANE	0.232	0.170	1.271	0.414	0.260	2.960	0.975	0.480	7.397	0.469	0.318	3.201
N-NONANE	0.033	0.026	0.187	0.057	0.050	0.270	0.079	0.048	0.434	0.469	0.361	1.607
N-OCTANE	0.046	0.037	0.191	0.100	0.080	0.440	0.107	0.072	0.720	0.091	0.078	0.244
N-PENTANE	0.877	0.762	2.383	0.508	0.480	1.280	0.751	0.626	3.083	0.936	0.657	7.593
N-PROPYLBENZENE	0.023	0.021	0.074	0.065	0.060	0.210	0.067	0.057	0.210	0.059	0.051	0.201
N-UNDECANE	0.033	0.030	0.136	0.011	0.000	0.190	0.094	0.073	0.396	0.133	0.115	0.291
O-ETHYLTOLUENE	0.024	0.021	0.086	0.058	0.050	0.200	0.078	0.057	0.322	0.067	0.055	0.263
O-XYLENE	0.175	0.126	0.933	0.256	0.200	1.270	1.058	0.633	8.043	0.327	0.256	1.176
P-DIETHYLBENZENE	Nan	Nan	Nan	0.000	0.000	0.000	0.071	0.050	0.563	0.054	0.042	0.151
P-ETHYLTOLUENE	0.030	0.025	0.127	0.043	0.040	0.160	0.107	0.076	0.478	0.086	0.070	0.329
PROPANE	3.651	3.456	11.666	2.355	2.130	9.360	4.801	3.754	20.957	1.221	1.121	2.860
PROPENE	0.581	0.496	2.472	0.897	0.870	3.430	0.568	0.358	3.387	0.785	0.717	2.336

STYRENE	0.040	0.026	0.383	0.202	0.170	1.270	0.180	0.079	2.078	0.119	0.084	0.493
TOLUENE	1.319	1.055	6.400	0.867	0.550	5.290	5.312	3.041	39.897	1.154	0.963	3.594
TRANS-2-BUTENE	0.110	0.092	0.470	0.003	0.000	0.260	0.201	0.186	0.395	0.175	0.125	1.851
TRANS-2-PENTENE	0.008	0.003	0.136	0.024	0.000	0.860	0.026	0.011	0.355	0.052	0.008	0.771

In addition, I do not know what NO2 chemical conversion to NO means as stated on line 25 of page 3. This needs to be described and the probability of interference from PAN needs to be discussed. I would expect at least 5 ppbv of PAN in areas such as Beijing during the day. This could lead to a significant interference in NO2. Please also report in more detail on the VOC observations. I think at least averages of the top 10 or 5 VOC in terms of OH loss should be listed for each city. I like the graphs in the supplement but the scales on many of the graphs don't make sense. Often the parameter graphed only goes up to 10 or 20% of full scale making it impossible to see what is going on. I don't think keeping consistent axes between different cities is worth not being able to read the graph.

Answer:

We changed the sentence to be "… after chemical conversion to NO." to be "in the form of NO by chemical conversion using Molybdenum convertor. This conversion method is known to be interference by NO_z species, which could be converted to NO. Therefore, one should keep in mind that the NO_2 measurement presented in this study could be positive biased from the ambient NO_2 concentrations." Besides, we derived the PAN concentrations from our box model calculations, which are about 2 ppbv (Fig. S7).

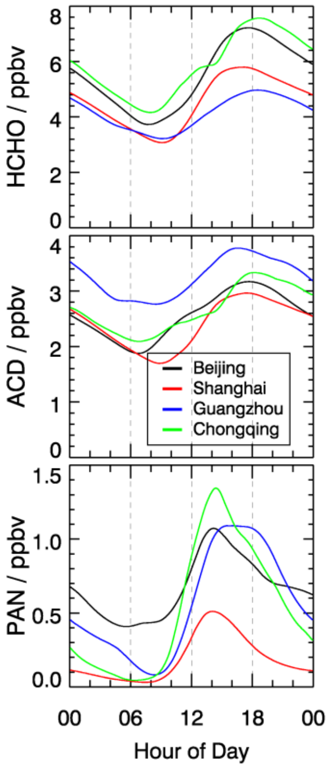

Figure S7. The mean diurnal profiles of modelled formaldehyde (HCHO), acetaldehyde (ACD) and peroxyacetyl nitrate (PAN) concentrations in four cities.

We added a table about measured VOC in supplement (Table S3) and a table showing top 10 k_{OH} contributing VOCs (Table 2). The mean diurnal profiles of top 10 VOCs are added in supplement (Fig. S6). A detail discussion on the measured VOCs is added. Please find the answers in the response to referee #1 who has the similar comments.

We changed the scale of the Fig. S1-S4 as suggested (see below).

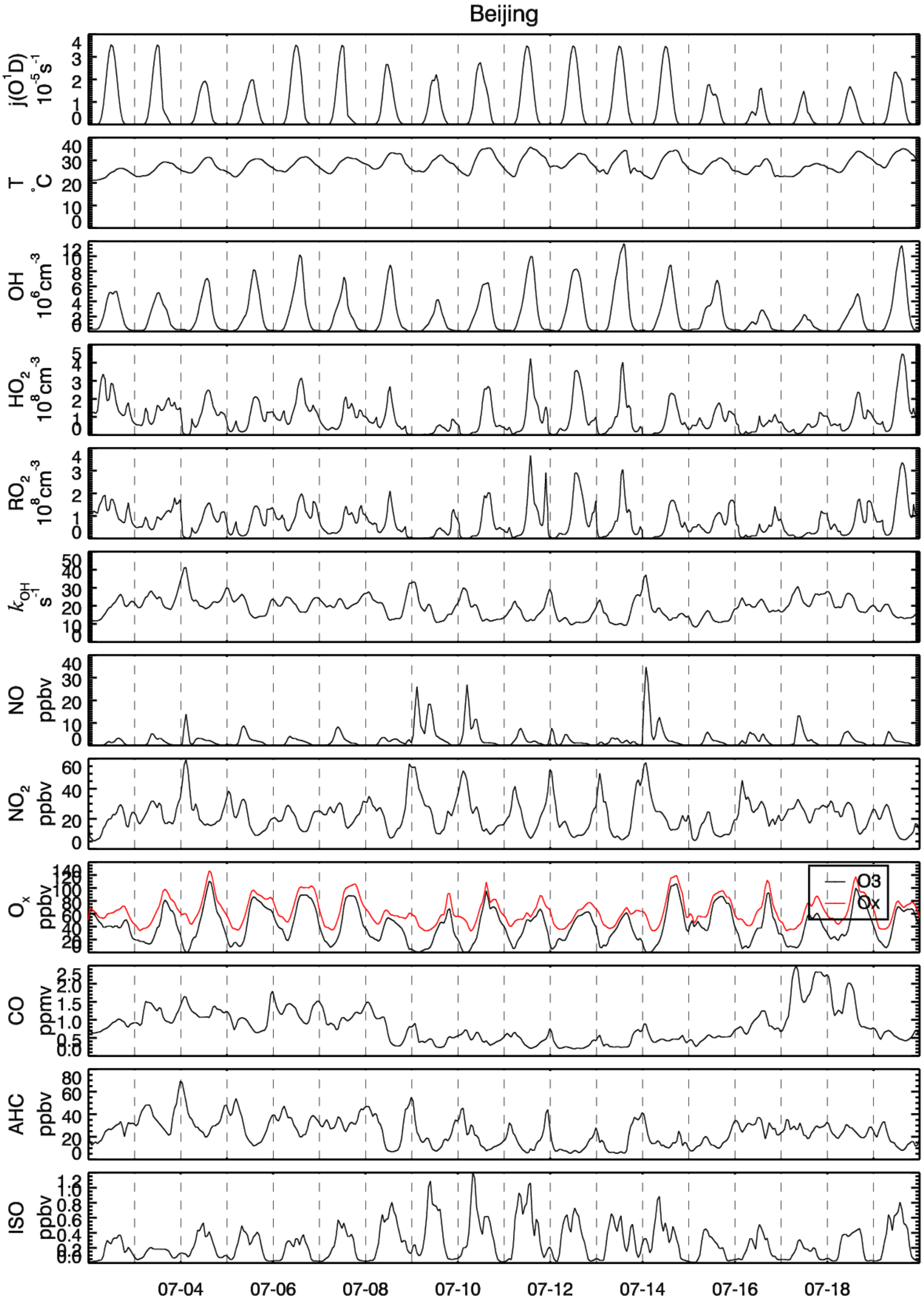

Figure S1 The time series of measured parameters (j(O^1D), Temperature, NO, NO_2, O_3, O_x, CO, AHC, isoprene) and modelled OH, HO_2, and RO_2 concentrations and OH reactivity in Beijing.

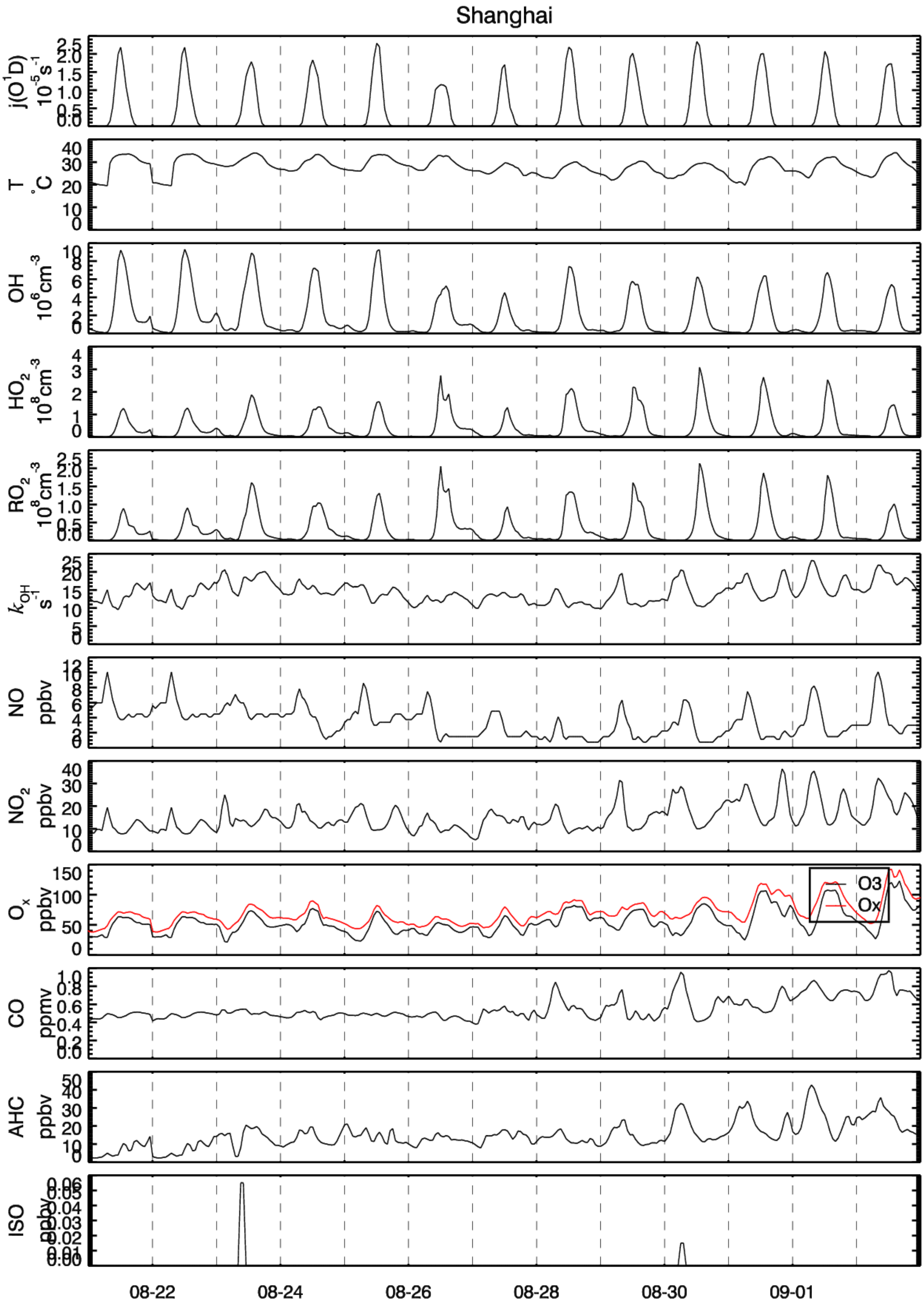

Figure S2 The time series of measured parameters (j(O^1D), Temperature, NO, NO_2, O_3, O_x, CO, AHC, isoprene) and modelled OH, HO_2, and RO_2 concentrations and OH reactivity in Shanghai.

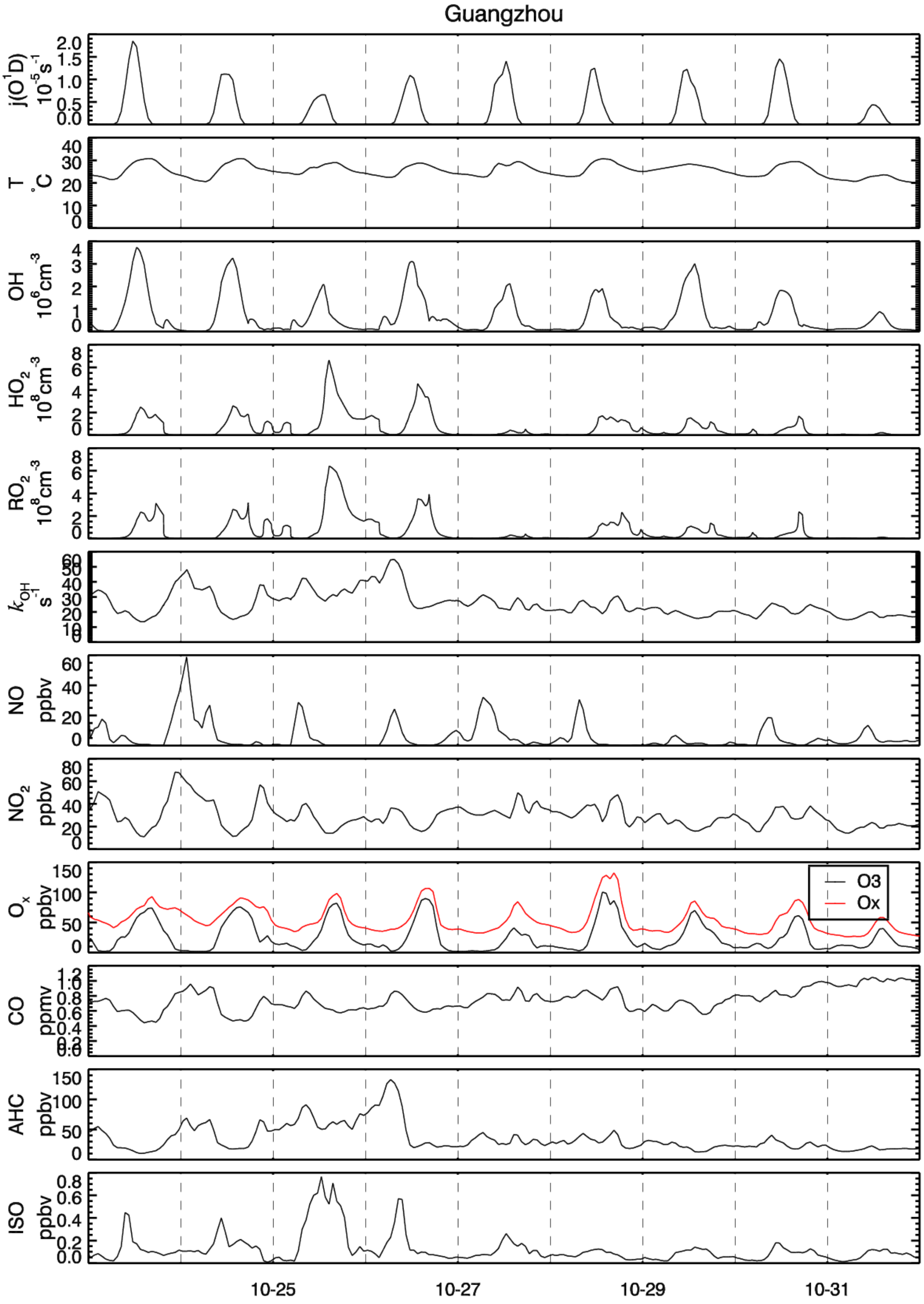

Figure S3 The time series of measured parameters (j(O^1D), Temperature, NO, NO_2, O_3, O_x, CO, AHC, isoprene) and modelled OH, HO_2, and RO_2 concentrations and OH reactivity in Guangzhou.

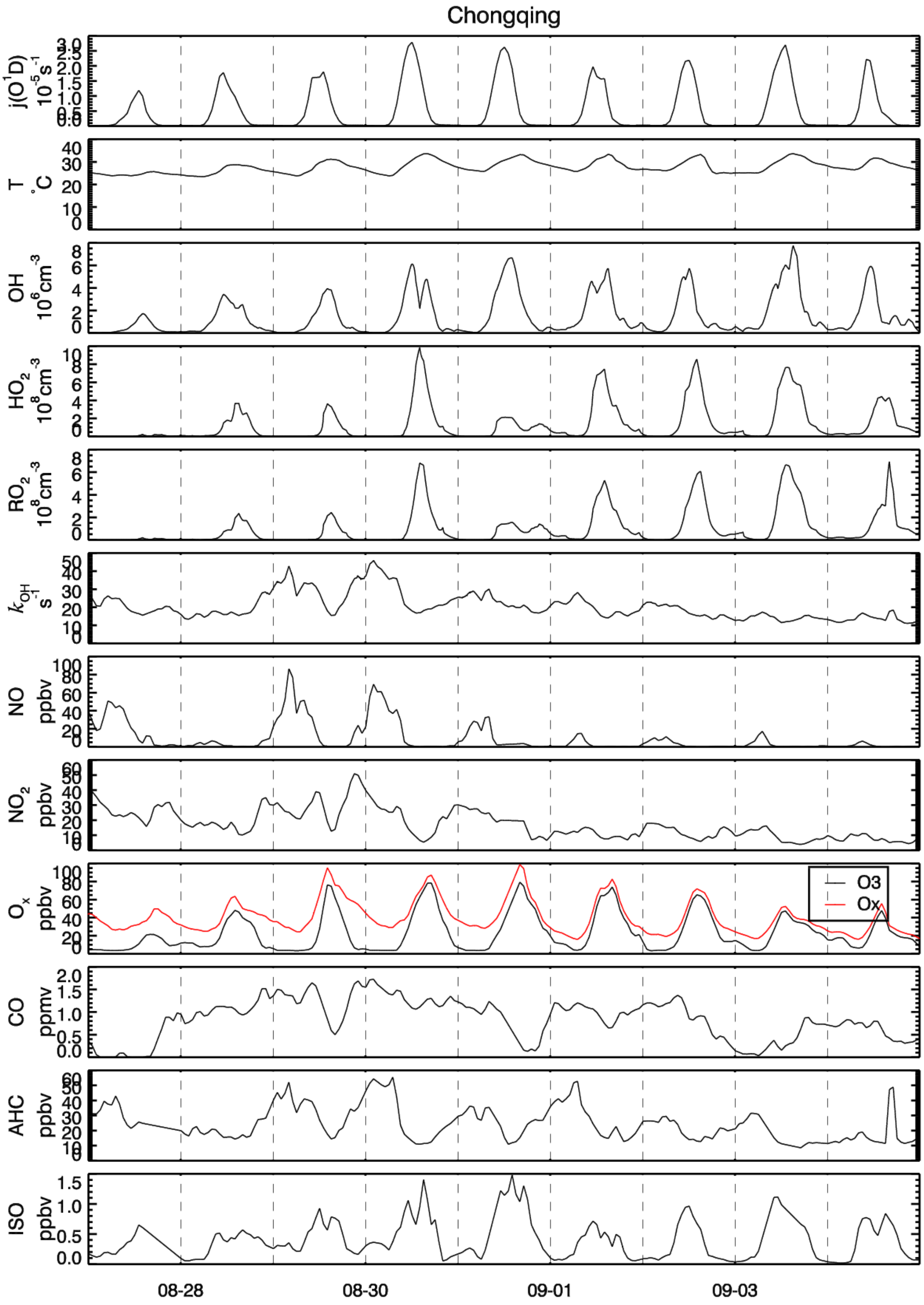

Figure S4 The time series of measured parameters (j(O^1D), Temperature, NO, NO_2, O_3, O_x, CO, AHC, isoprene) and modelled OH, HO_2, and RO_2 concentrations and OH reactivity in Chongqing.

3) The reporting of OH reactivity could be made much simpler as well. Perhaps having a *section in the results showing the VOC observations separately would be less confusing. You could then have a following section on the calculated OH reactivity. I really recommend limiting the discussion in these sections and focusing on the results. For example, the paragraph on line 1 page 5 stating that OH reactivity can be measured in 3 ways made me think for some time that this was a measured quantity in this work.*

Answer:

We removed the part of the description of OH reactivity measurement techniques. The description of VOC measurement and discussion are moved to section 4.1. Please see detail in the response to referee#1.

4) I highly recommend being more explicit on what is derived from the model or parameterized. For example, I don't think formaldehyde or acetaldehyde are measured but are model predicted. If so this needs to be described and predicted levels compared to observations if available. This will certainly impact the radical budget as well as the production rate of PAN relative to HNO3. So I suggest a table of model parameters that are predicted, constrained, and parameterized. I also suggest that the model results be presented in an organized manner in the results section. There is a lot of discussion throughout section 3 that should probably be in section 4.

Answer:

Similar to the comments (2), we prepared the table in supplement to state what are measured, modelled, and parameterized in this study (Table S2). We added a sentence in the new section 4.1 to discuss the modeled OVOC concentration "The OVOCs concentrations are simulated by the box model. The modelled HCHO concentrations were in the range of 3 to 8 ppbv (Fig. S7), which are consistent with the previous studies in these regions (Zhang et al., 2012;Song et al., 2018;Chen et al., 2016;Tang et al., 2009). The modelled acetaldehyde concentrations are in the range of 2 to 3 ppbv in Beijing, Shanghai, and Chongqing but on average 1 ppbv larger in Guangzhou because the larger contribution of aromatics VOCs which produce acetaldehyde from their OH degradation."

We restructured the manuscript by moving the VOC description to section 4.1 and moving the OH-HO_2-RO_2 budget analysis (originally section 3.3.2) to section 4.2.

5) The very simple parameterization of HONO as being 2% of NO2 is somewhat troubling. I am surprised that it would be that simple especially as a function of the time of day. I think this assumption needs to be better justified and probably looked at to determine the sensitivity, i.e. some case studies with different assumptions are probably needed. This is also another reason to describe the NO2 measurement in more detail.

Answer:

Referee #1 has similar concern on the uncertainty in HONO parameterization. We performed more sensitivity study to investigate the uncertainty and please find our answer in the response to Referee#1.

6) I am not sure the ISOROPPIA modeling adds much to the paper especially as there are no measurements of ammonia or nitric acid. I certainly realize that if there is a large excess of ammonia that this will drive nitric acid into the aerosol. However, I am not sure the nitric formation rate vs. loss rate to aerosol versus dry and wet deposition can be suitably treated in this work to allow for quantitative predictions of ammonium nitrate aerosol. So I recommend removing from the paper and perhaps replacing with a simple discussion. This discussion could also mention that cutting down NOx may lead to enhanced ozone production but it will cut down on particulate nitrate as well.

Answer:

We agree that the calculation may not be quantitative since some key parameters, e.g. ammonia and nitric acid concentrations were not measured during these campaigns. We reduce the content in the section Nitrate production potential. We also moved some of the contents and original Table 2 to supplement. Please find the changes in the main text following.

“Nitric acid is one of the major products generated by the radical system for high NO_x conditions, which is an important precursor of particulate nitrate (NO_3^-). Recently, nitrate has become a significant portion in particles in Beijing, Shanghai, and Nanjing (YRD) during summertime in China (Li et al., 2018). The gas phase nitric acid HNO_3 together with ammonium NH_3 form a gas-particle partitioning equilibrium with NH_4NO_3 (R3), which depends on the relative humidity, temperature, and the aerosol contents (Seinfeld and Pandis, 2016).

$$\mathrm{NH_3} + HNO_3 \leftrightarrow NH_4NO_3 \qquad \text{(R3)}$$

The nitric acid is mainly produced from the reaction between NO and OH which can be derived from the box model. The fate of nitric acid depends on the gas-particle partition, deposition and chemical reactions. In general, the time scale of partitioning is 1-2 orders smaller than those of deposition and chemical production (Morino et al., 2006;Neuman et al., 2003). Therefore, the photochemical produced nitric acid will deposit on to the aerosol if the ambient NH_3 is sufficient. The deposition rate is about 7 cm s^{-1} (Seinfeld and Pandis, 2016), which results in a deposition timescale being 8 hours if the boundary layer height is 2 km (typical values for summertime). The ammonia concentrations are usually above 10 $\mu g/m^3$ in urban areas in China during summertime (Pan et al., 2018), which indicates ammonia-rich conditions and sufficient to neutralize nitric acid..

In this study, we use the aerosol thermodynamic model (ISORROPIA) to simulate the nitrate production and the model design is explained in supplement. It’s worth noting that such model simulation cannot be quantitatively because some key parameters, e.g. ammonia and nitric acid concentrations were not measured during these campaigns. The discussion below should be considered as a qualitative estimation to show the important feature in determining the particle nitrate production. The modeled nitrate concentration and partitioning in Beijing are shown to illustrate the typical pattern of particulate nitrate formation (Fig. 11a). The total nitrate concentrations maximize in the late afternoon while the particulate nitrate shows a board peak at night, which is mainly driven by the stronger gas-to-particle partitioning due to higher RH. Since deliquesce relative humidity (DRH) of NH_4NO_3 is about 60% in all cases, the partitioning changed dramatically with the relative humidity above DRH (nighttime) and below DRH (daytime). One should note that the nitrate formation from N_2O_5 hydrolysis is not taken into account in this study, which could lead to negative bias in the nitrate production calculation.

To investigate the nitrate concentration dependence on the nitrate production rate and ambient ammonia concentrations, the averaged nitrate concentrations are plotted as a function of daily integrated nitric acid production rate and total ammonium ($NH_4^+{}_{(a)}+NH_{3(g)}$) concentrations. As shown in Fig. 11b, the isopleth diagrams are split into two parts by the dashed line to represent the nitrate- (upper left) and ammonium-sensitive (lower right) regimes. However, the threshold for nitrate- and the ammonium-sensitive regime is not distinct in the small chemical range. Actually, the nitrate concentrations are sensitive to both precursors. The daily integrated nitrate production rate and averaged total ammonium concentrations for each city are denoted by the circles (Fig. 11b). The circles are located above the ridgeline, which means nitrate concentrations are more sensitive to the change of nitric acid production rate. Therefore, this scenario study highlights that the further mitigation of summertime particulate nitrate pollution should aim at the reduction of photochemical nitric acid production. For example, the reduction in NO_x emission could help to reduce the particulate nitrate pollution but may lead to enhancement in ozone pollution (see section 4.3). ”

Reference:

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