

Dear Editor and Reviewers,

We would like to thank Referee #2 for valuable comments on our manuscript. We have carefully revised our manuscript according to the referee's comments. We have addressed the referee's comments and concerns point by point as follows (comments from referee in black, author's response in red and author's changes in the revised manuscript in blue):

Comment:

The authors estimated chlorine emissions from coal combustion and prescribed waste incineration in China and used the CMAQ model to examine the impact of these chlorine emissions on ozone. Overall, the article is written clearly and merits publication. However, several issues need to be addressed before publication.

Responses:

We thank the referee for his nice comments.

Changes in the manuscript:

No changes were needed for this comment.

Comment:

Page 2, line 16-17, equation 1-2 The authors may replace "H" in both reactions with "h".

Responses:

We have corrected the typo.

Changes in the manuscript:

We have corrected it in line 16-17 on page 2 in the revised manuscript.

Comment:

Page 6, line 19-21 Incomplete sentence

Responses:

We have revised this sentence to "Table 1 also lists the waste incineration from garbage disposal incinerators in each province/city from China Urban-Rural Construction Statistical Yearbook".

Changes in the manuscript:

We have corrected it in line 26-28 on page 6 in the revised manuscript.

Comment:

Page 7, line 19-29 How the chlorine emissions were temporally allocated?

Responses:

To include the ACEIC in the CMAQ model, the chlorine emissions from different economic sectors were temporally allocated in different ways. For the coal combustion from the power plant, industrial and residential sectors, we distributed the total chlorine emissions into each month according to Wu (2009). The monthly variations of chlorine emissions from each sector are shown in Fig. R1. In addition, the daily distributions of chlorine emissions from the power plant, industrial and residential sectors were allocated the same way as the allocations of the MIX inventory from the corresponding sectors developed by Tsinghua University (<http://www.meicmodel.org>). For the coal combustion from other sector, the total chlorine emission was divided equally into each month, each day and each hour.

Since the burning process of garbage disposal incinerators is similar to that of the power plants, we assumed the same monthly and daily variation of prescribed waste incineration as that of the power plant sector.

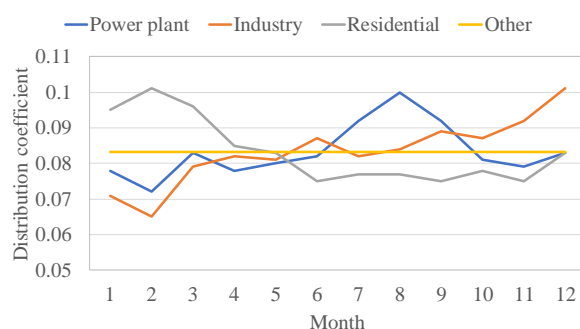


Figure R1 Monthly distributions of chlorine emissions from the power plant, industrial, residential and other sectors.

#### References:

Wu, X. L.: The study of air pollution emission inventory in Yangtze Delta, M.S. thesis, Fudan University, China, 94 pp., 2009.

#### Changes in the manuscript:

The above statements have been added in line 22-29 on page 9 in the revised manuscript. We also added the reference (Wu, 2009) in line 22-23 on page 18.

#### Comment:

Page 9, line 13-15 ACEIC has been defined before. It appears that ACEIC has been defined several times throughout the article. Please check and define it once.

#### Responses:

We have corrected this redundancy. It should read only once now.

#### Changes in the manuscript:

We have corrected it in line 20-21 on page 9 and line 14-15 on page 13 in the revised manuscript.

#### Comment:

Page 11, line 1-10 The authors reported that chlorine emissions/chemistry increased the conversion of  $\text{NH}_3$  into  $\text{NH}_4^+$ . What mechanism caused the increased conversion of  $\text{NH}_3$  into  $\text{NH}_4^+$ ?

#### Responses:

Volatile acidic species (i.e., HCl) can be partitioned into particles by neutralization reactions (Seinfeld and Pandis, 1998). Semi-volatile  $\text{NH}_4\text{Cl}$  is formed via reversible phase equilibrium with  $\text{NH}_3$  and HCl (Pio and Harrison, 1987). When the HCl emission was included in the model, HCl reacts with  $\text{NH}_3$  to produce particulate  $\text{NH}_4^+$  and  $\text{Cl}^-$ , provided that the  $\text{NH}_3$  emission was sufficiently high.

#### References:

Pio, C. A., and Harrison, R. M.: Vapour pressure of ammonium chloride aerosol: effect of

temperature and humidity, *Atmos. Environ.*, 21(12), 2711–2715, doi:10.1016/0004-6981(87)90203-4, 1987.

Seinfeld, J. H., and Pandis, S. N.: *Atmospheric Chemistry and Physics*, John Wiley & Sons, Inc., New York, 1998.

Changes in the manuscript:

The above statements have been added in line 12-21 on page 11 in the revised manuscript.

Comment:

Page 12, line 9-21 The authors demonstrated the impact of chlorine emissions/chemistry on daily maximum 1-hr O<sub>3</sub>. Chlorine emissions/chemistry tends to increase ozone in the morning hours. Thus, it is likely to have a larger impact on 8-hr O<sub>3</sub> on than on 1-hr O<sub>3</sub>. It will be important to present impact of chlorine emissions/chemistry on 8-hr O<sub>3</sub> also.

Responses:

Figure R2 presents the impact of chlorine emissions on 8-h O<sub>3</sub>. The inclusion of chlorine emission in the CMAQ model increased the monthly mean daily maximum 8-h O<sub>3</sub> by up to 2.0 ppbv (4.1%), similar to the impact of chlorine emission on 1-h O<sub>3</sub>. It is reasonable that 8-h O<sub>3</sub> is more representative than 1-h O<sub>3</sub>. Hence, we moved the results of 1-h O<sub>3</sub> to the supplementary (Fig. S9a-c) and included that of 8-h O<sub>3</sub> in the manuscript (Fig. 8a-c).

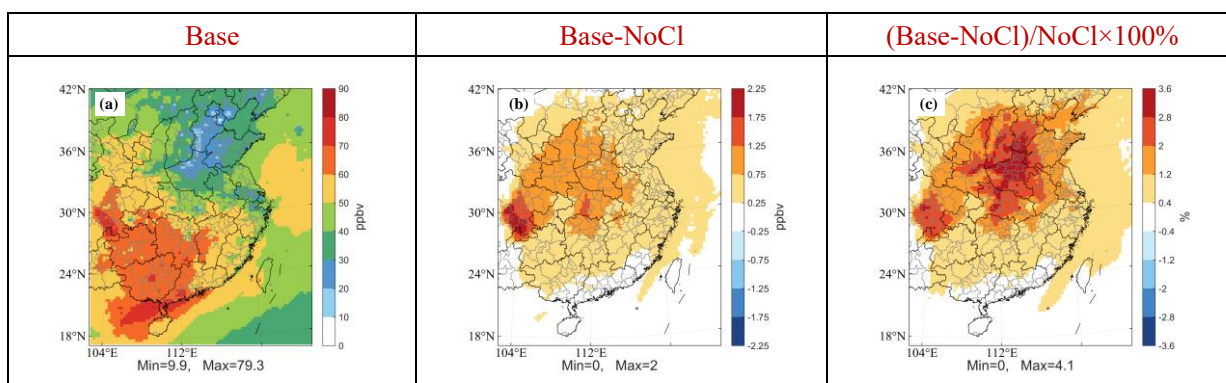


Figure R2 Comparison of the monthly mean concentrations of daily maximum 8-h O<sub>3</sub> in the Base experiment, the differences (Base minus NoCl), and the percent changes to the NoCl experiment.

Changes in the manuscript:

The above statements have been added in line 1-5 on page 13 in the revised manuscript. We placed Fig. R2a-c (the results of 8-h O<sub>3</sub>) to Fig. 8a-c in the revised manuscript and moved Fig. 8d-f (the results of 1-h O<sub>3</sub>) in the original manuscript to the supplementary as Fig. S9a-c. Besides, the results of 1-h O<sub>3</sub> in Abstract (line 27 on page 1) and Conclusions (line 27-28 on page 13) were replaced with that of 8-h O<sub>3</sub>.

Comment:

Page 13, line 1-10 The authors reported that chlorine emissions/chemistry decreased NO<sub>x</sub> concentrations without providing any explanation? Why NO<sub>x</sub> concentration decreased?

Responses:

The increase of Cl radical concentration can enhance the atmospheric oxidation, leading to the

increase of ozone and OH radical concentrations. The OH radicals can in turn oxidize  $\text{NO}_x$  to produce particulate  $\text{NO}_3^-$ , resulting in the slight decrease of  $\text{NO}_x$  concentration.

Changes in the manuscript:

The above statements have been added in line 29-31 on page 12 and line 1-10 on page 13 in the revised manuscript. Also, we placed Fig. 8a-c (the results of  $\text{NO}_x$ ) in the original manuscript to Fig. 8d-f in the revised manuscript.

Comment:

Figure 1 Unit of chlorine emissions is written as Mg/grid; should it be written as Mg/grid/yr.

Responses:

We have revised “Mg grid<sup>-1</sup>” to “Mg grid<sup>-1</sup> yr<sup>-1</sup>” in Figure 1, S1, S2 and S3.

Changes in manuscript:

We have corrected it in Fig. 1 in the revised manuscript and Fig. S1, S2 and S3 in the revised supplementary.

Comment:

Figure 2 Figure title indicates that fractional changes are shown. Actually percent changes are shown.

Responses:

We have revised “fraction change” to “percent changes” in Fig. 4 and 8.

Changes in manuscript:

We have corrected it in Fig. 4 and 8 in the revised manuscript.

Comment:

Figure S2 It contains four captions: (a-d). However, (c) is written twice, one should be written as (d).

Responses:

We have corrected this typo.

Changes in the manuscript:

We have corrected it in Fig. S2 in the revised supplementary.