

### General comments:

This paper (acp-2014-547) reported an experimental study on the heterogeneous reaction between OH radicals and TBEP using particle-phase relative rates technique. The second-order rate constant for the reaction was obtained and the atmospheric lifetimes of TBEP and other OPEs were estimated. The influence of mixing state of multi-component particle on TBEP oxidation was also investigated. TBEP is a widely used flame retardant, which can be emitted in the atmosphere primarily in the particle phase. The results reported in this paper are important for assessing the persistence of TBEP and its fate in the atmosphere. This paper matches the merit of Atmospheric Chemistry and Physics. However, there are still some problems and mistakes that need to be addressed prior to possible publication.

(1) The usage of equation (Eq. 5) for calculating uptake coefficient  $\gamma_{OH}$  is questionable. First, the second-order reaction constant  $k_{2,TBEP}$  should be in the numerator other than denominator. Second, this equation is appropriate for calculating the  $\gamma_{OH}$  involving pure particles other than mixed particles. According to the original author Smith et al., derivation of  $\gamma_{OH}$  is as follows:

$$\gamma_{OH} = \frac{-d[TBEP]/dt}{f \cdot J_{coll} \cdot A \cdot C_p}$$

Where  $f$  is the fraction of TBEP molecules remaining within the particle (i. e.  $[TBEP]/[TBEP]_0$ );  $J_{coll}$  is the OH flux at the particle surface ( $\frac{1}{4} \bar{c}[OH]$ );  $A$  is the particle surface area, and  $C_p$  is the particle number concentration. Then, the  $\gamma_{OH}$  is,

$$\gamma_{OH} = \frac{k_{2,TBEP}[TBEP][OH]}{\frac{[TBEP]}{[TBEP]_0} \cdot J_{coll} \cdot A \cdot C_p}$$

$$= \frac{k_{2,TBEP}[TBEP]_0[OH]}{J_{coll} \cdot A \cdot C_p}$$

Here, if the TBEP particles used in this experiment are single-component pure TBEP particles, like the particles used in the study of Smith et al, the initial concentration of TBEP ( $[TBEP]_0$ , in molec cm<sup>-3</sup>) can be calculated in terms of the initial particle-phase density  $\rho_{TBEP}$ .

$$[TBEP]_0 = \frac{C_p \cdot V \cdot \rho_{TBEP} \cdot N_A}{M_{TBEP}}$$

Where V is the particle volume,  $M_{TBEP}$  is the molar mass of TBEP,  $N_A$  is Avogadro's number. Put it in the above equation of  $\gamma_{OH}$ ,

$$\gamma_{OH} = \frac{k_{2,TBEP} \frac{C_p \cdot V \cdot \rho_{TBEP} \cdot N_A}{M_{TBEP}} [OH]}{\frac{1}{4} \bar{c}[OH] \cdot A \cdot C_p}$$

Since  $V/A$  equals to  $\frac{D_p}{6}$  ( $D_p$  is the surface-weighted particle diameter), the final expression for  $\gamma_{OH}$  should be,

$$\gamma_{OH} = \frac{2k_{2,TBEP} D_p \rho_{TBEP} N_A}{3\bar{c} M_{TBEP}}$$

To sum up,  $\rho_{TBEP}$  in this equation to calculate the initial concentration of TBEP ( $[TBEP]_0$ ) is improper. The obtained  $[TBEP]_0$  is the initial concentration of pure TBEP particles. However, in fact, a fraction of particles is occupied by other substances such as ammonium nitrate in this experiment. Thus, the exact value of  $[TBEP]_0$  must be not accordance with the calculated value of this study.

(2) Page 16, Lines 4-6: The author states that “the diffusion rate of CA or OH radicals in the particle phase increases at higher RH, subsequently enhancing the potential for reaction with OH”. However, from Figure 7(c) we can see that the consumption of CA under higher RH was less than that under lower RH. These experimental results do not support the conclusion.

**Minor comments:**

Page 2, Line 20: Show the full name of “PM” for its first appearance in the article.

Page 8: Since PMF analysis is not a widely-used method in data processing, a brief description for its principle can be added in the Section 2.2. For example,  $Q/Q_{\text{exp}}$  (Page 10, Line 1) first appears in the paper but without an explanation.

Page 10, Line 20: “a small fraction factor 4” may be “a small fraction of factor 4”.

Page 10, Line 25: “227 and 299” should be “227, and 299”.

Page 11, Line 2: “Figure 3a-d further compares...” should be “Figures 3a-d further compare...”.

Page 11, Line 9: Add comma “,” after “227”.

Page 11, Line 11: Replace “citric acid” by “CA”.

Page 11, Line 14: Add comma “,” after “oxidized”.

Page 11, Line 15: “is” should be “are”.

Page 11, Lines 16 and 17: “Fig. S3a-c” should be “Figs. S3a-c”. Please unify the expression of Figure in the full text. For example, the full names “Figures 1 and 2”

and “Figure 3a-d” were used in the Page 9, Line 24 and Page 11, Line 2, but the abbreviation “Fig. S3a-c” was used here and other places.

Page 11, Line 24: Substitute the oral language “In doing so” by some written words.

Page 11, Line 27 and Page 12, Line 1: The present tense and past tense are in one sentence. Please revise it.

Page 12, Lines 8-10: The same problem as above.

Page 13, Lines 23, 25, 27...: The units of rate constants for many places in this paper are incorrect. “ $\text{cm}^3 \text{ molecule s}^{-1}$ ” should be “ $\text{cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ ”.

Page 14, Line 17: “Fig. 6a and b” should be “Figs. 6a and b”. Please use the plural form of Fig., if necessary, for other places throughout the article.

Page 17, Line 1: The “ $10^{-1}$ ” should be “ $10^{-10}$ ”.

Page 17, Line 22: Add comma “,” after “TEHP”. There are still many similar problems in this paper. Please revise the format “A, B and C” with “A, B, and C”.

Page 20, Lines 10-13: There is a grammatical mistake in this sentence: “*In particular, the measurements of TPhP in PM in remote regions (Möller et al., 2012), despite its dominant gas-phase loss contribution (based upon our model results) highlights the effect of multi-component particle mixtures on the kinetics of particle degradation.*”

Page 20, Line 22: Delete the word “show” after “that”.

Page 31, the caption of Figure 1: Replace “Exp 1, 2, 3 represent...” by “Error bars represent...”.

Page 37, the caption of Figure 7: Please state clearly that the RH of experiments (A) and (B) are  $(30 \pm 3)\%$ , and that of (C) is  $(57 \pm 2)\%$ ”.

