Review for "Uncertainties in the effects of organic aerosol coatings on polycyclic aromatic hydrocarbon concentrations and their estimated health effects" by Lou et al.

In this study the authors compared three BaP degradation approaches with different treatment of the effects of organic aerosol coatings on the particle-phase BaP reactivity. The simulated gas-phase BaP and oxidized BaP based on the three approaches were compared with observations. As BaP has significant health effects, accurate representation of BaP concentrations over a global scale is highly needed. This manuscript is clearly written and is helpful for future modelling studies to improve the PAHs simulations. I recommend the publication of this work after the authors could address my comments below.

Major comments:

(1) My major concern is that how to simulate the effects of organic aerosol coating, namely, the viscosity, on the gas-particle partitioning of BaP and the heterogeneous degradation coherently. Viscosity affects both partitioning and reactivity. Therefore, if the Bap degradation in the particle phase has considered the effects of viscosity in the Shielded approach and ROI-T approach as what the author did, for coherence, the effect of viscosity on gas-particle partitioning of BaP should be also included. Does the ppLFER model the authors applied for gas-particle partitioning of BaP specifically considered the viscosity of the organic shell and its effect on partitioning?

(2) My another concern is that the section of "Conclusion and Discussion" is too much rephrasing the results with little discussion. I recommend that the authors can give a clearer conclusion. For the discussion, the following aspects may be helpful. First, what are the limitations of the Shielded and ROI-T approaches and how to improve these kinds of methods for future modelling studies? For example, Bap and ROI-T have not considered the phase state of the organic coatings varied with RH, *T*, and organic aerosol chemical composition, which, however, has been parameterized since 2017 (Li et al., 2020; Shiraiwa et al., 2017; Zhang et al., 2019) and included in chemical transport models (Shrivastava et al., 2022; Zhang et al., 2024). Second, would phase phase separation affect BaP gas-particle partitioning and heterogeneous degradation (Pye et al., 2017; Schmedding et al., 2020)?

Specific comments:

(1) Some recent modeling work on PAH should be cited and the simulated BaP concentrations are better compared (Wu et al., 2024).

(2) Line 164 at Page 5, why the authors did not include the gas-phase reaction of BaP with O_3 and NO_3?

(3) Line 348-350 at Page 11, the authors wrote that a large portion of the total Bap was still oxidized though the effect of organic coating had been considered in bulk diffusion of BaP in the ROI-T scheme, which was surprising. Again, referring to my major comment 1, the authors should clarify that the effect of viscosity on

gas-particle partitioning and BaP degradation rate is related to two different processes. Did the ROI-T scheme consider the phase state effect on bulk diffusivity of BaP in your simulation? I think the result showed in Line 348-350 is not surprising as the reaction rate of BaP with ozone in the particle phase was dependent on RH and T, thus the viscosity of organic shell, which resulted in limited effect of organic coating on BaP degradation in areas with low viscous particles.

(4) Line 198-200 on Page 6, please double check. I did not see this from Table S3 in Mu et al. (2018).

References

1. Li, Y., Day, D. A., Stark, H., Jimenez, J. L., and Shiraiwa, M.: Predictions of the glass transition temperature and viscosity of organic aerosols from volatility distributions, Atmos. Chem. Phys., 20, 8103-8122, 10.5194/acp-20-8103-2020, 2020.
2. Pye, H. O. T., Murphy, B. N., Xu, L., Ng, N. L., Carlton, A. G., Guo, H., Weber, R., Vasilakos, P., Appel, K. W., Budisulistiorini, S. H., Surratt, J. D., Nenes, A., Hu, W., Jimenez, J. L., Isaacman-VanWertz, G., Misztal, P. K., and Goldstein, A. H.: On the implications of aerosol liquid water and phase separation for organic aerosol mass, Atmos. Chem. Phys., 17, 343-369, 10.5194/acp-17-343-2017, 2017.
3. Schmedding, R., Rasool, Q. Z., Zhang, Y., Pye, H. O. T., Zhang, H., Chen, Y., Surratt, J. D., Lopez-Hilfiker, F. D., Thornton, J. A., Goldstein, A. H., and Vizuete, W.: Predicting secondary organic aerosol phase state and viscosity and its effect on multiphase chemistry in a regional-scale air quality model, Atmos. Chem. Phys., 20, 8201-8225, 10.5194/acp-20-8201-2020, 2020.
4. Shiraiwa, M., Li, Y., Tsimpidi, A. P., Karydis, V. A., Berkemeier, T., Pandis, S. N., Lelieveld, J., Koop, T., and Pöschl, U.: Global distribution of particle phase state in atmospheric secondary organic aerosols, Nat. Commun., 8, 15002, 10.1038/ncomms15002, 2017.
5. Shrivastava, M., Rasool, Q. Z., Zhao, B., Octaviani, M., Zaveri, R. A., Zelenyuk, A., Gaudet, B., Liu, Y., Shilling, J. E., Schneider, J., Schulz, C., Zöger, M., Martin, S. T., Ye, J., Guenther, A., Souza, R. F., Wendisch, M., and Pöschl, U.: Tight Coupling of Surface and In-Plant Biochemistry and Convection Governs Key Fine Particulate Components over the Amazon Rainforest, ACS Earth and Space Chemistry, 6, 380-390, 10.1021/acsearthspacechem.1c00356, 2022.
6. Wu, Z., Chen, X., Wang, Z., Chen, H., Wang, Z., Mu, Q., Wu, L., Wang, W., Tang, X., Li, J., Li, Y., Wu, Q., Wang, Y., Zou, Z., and Jiang, Z.: Modeling of polycyclic aromatic hydrocarbons (PAHs) from global to regional scales: model development (IAP-AACM_PAH v1.0) and investigation of health risks in 2013 and 2018 in China, Geosci. Model Dev., 17, 8885-8907, 10.5194/gmd-17-8885-2024, 2024.
7. Zhang, Y., Nichman, L., Spencer, P., Jung, J. I., Lee, A., Heffernan, B. K., Gold, A., Zhang, Z., Chen, Y., Canagaratna, M. R., Jayne, J. T., Worsnop, D. R., Onasch, T. B., Surratt, J. D., Chandler, D., Davidovits, P., and Kolb, C. E.: The Cooling Rate and Volatility Dependent Glass Forming Properties of Organic Aerosols Measured by Broadband Dielectric Spectroscopy, Environmental Science & Technology, 10.1021/acs.est.9b03317, 2019.
8. Zhang, Z., Li, Y., Ran, H., An, J., Qu, Y., Zhou, W., Xu, W., Hu, W., Xie, H., Wang, Z.,

Sun, Y., and Shiraiwa, M.: Simulated phase state and viscosity of secondary organic aerosols over China, Atmos. Chem. Phys., 24, 4809-4826, 10.5194/acp-24-4809-2024, 2024.