Supplement of Aerosol Research, 3, 205–218, 2025
https://doi.org/10.5194/ar-3-205-2025-supplement

AEROSOL Research
Open Access

Supplement of

Emission dynamics of reactive oxygen species and oxidative potential in particles from a petrol car and wood stove

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Supplement Information

Particle losses were characterized in a separate set of experiments using SOA produced via dark ozonolysis in a flow tube reactor. SOA was generated by passing 0.3 l/min of synthetic air over a heart-shaped flask filled with α-pinene. This flow was mixed with 4.7 l/min coming from an ozone-generating UV lamp (UVP LLC) into a 2.5 l glass flow tube. After the flow tube a dilution flow of 12 l/min was added. Similar particle mass and number concentrations as well as flow rates through the denuders were recreated as present during the emission measurements. By comparing the particle number size distributions measured with a scanning mobility particle sizer (TSI) before and after the denuders, the particle losses were determined to be approximately 10%

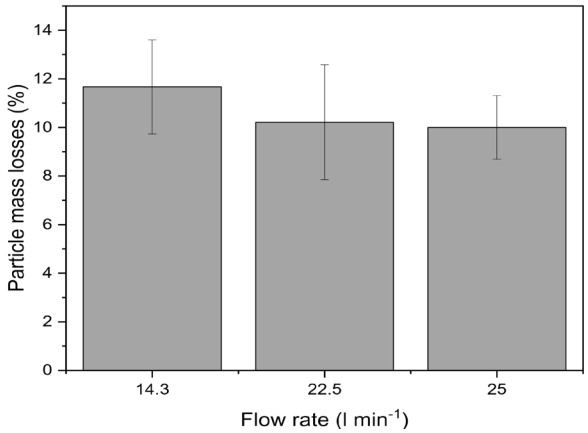

Figure S 1: Particle loss caused by the charcoal denuders at different flow rates

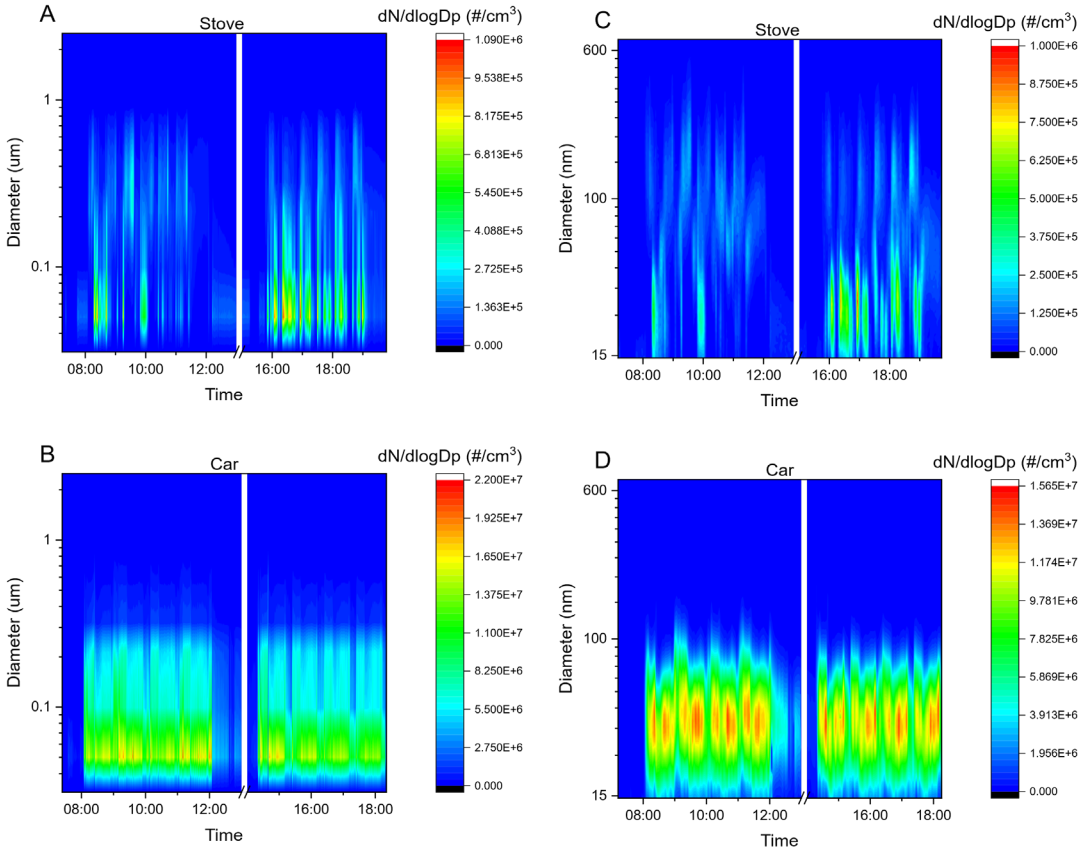

Figure S 2: ELPI contour plots of particle numbers for one stove (A) and one car (B) experiment. No significant particle number concentrations were measured in higher size bins. Equivalent SMPS contour plots of the same stove (C) and car (D) measurements show the absence of particles above 600 nm for stove and above 200nm for car emissions, respectively.

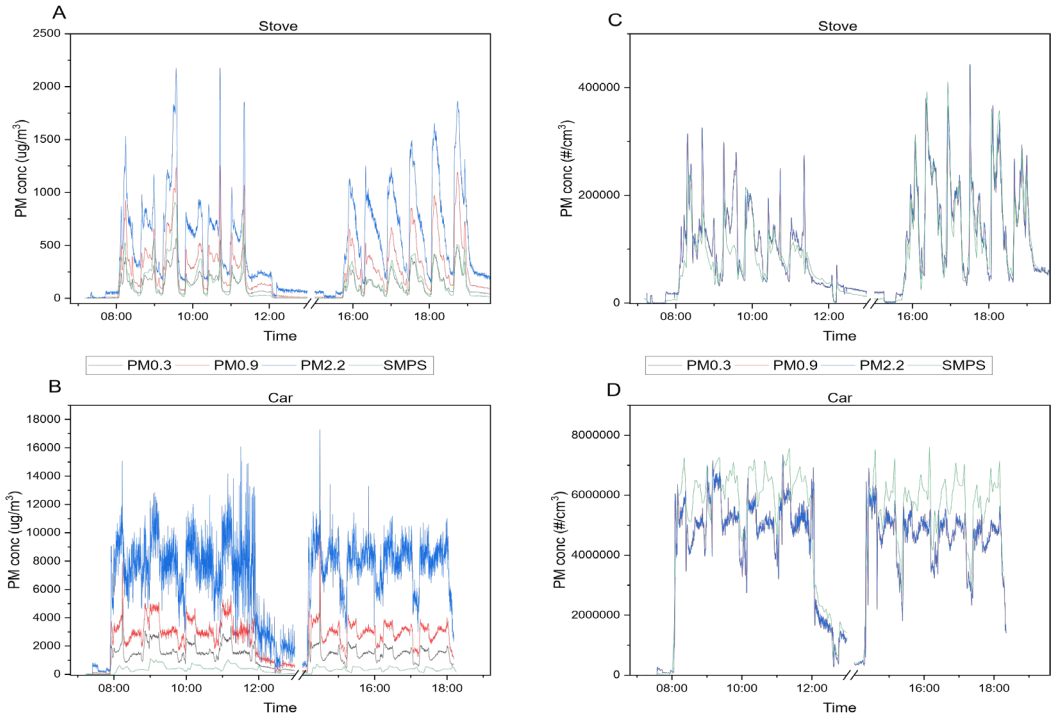

Figure S 3: Mass concentrations measured by the SMPS and ELPI during a stove (A) and car (B) exhaust measurement. The ELPI results are shown for three size ranges. Number concentrations measured by the SMPS and ELPI during a stove (C) and car (D) measurement. The ELPI results are shown for three size ranges. Nearly no number concentration differences between the different size ranges of the ELPI are visible. However, large differences in mass concentrations were caused by few artefact particles in larger size bins.

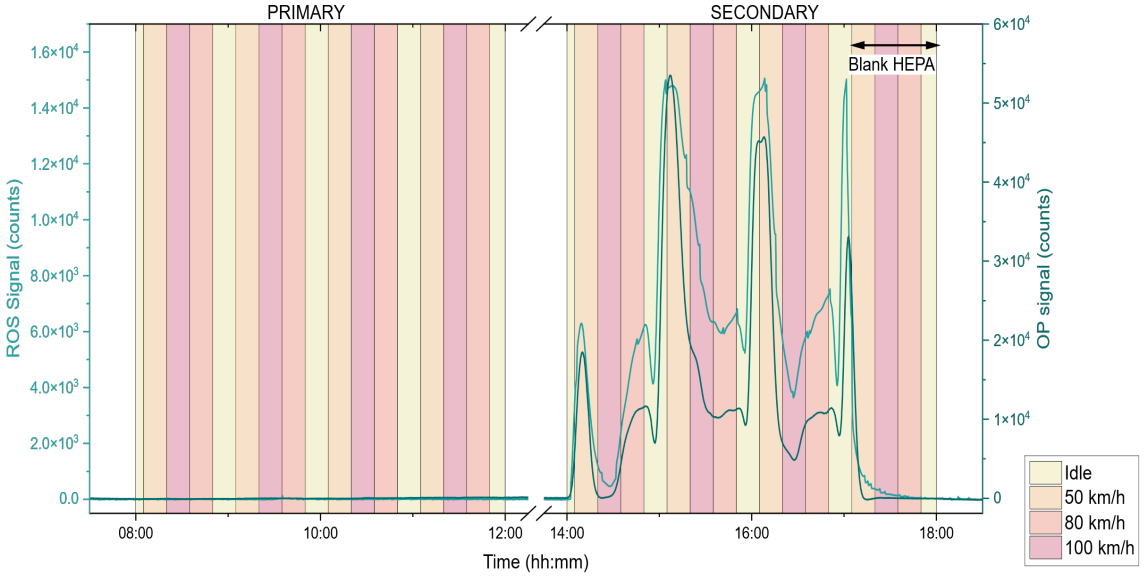

Figure S 4: Blank-corrected signal intensities of OP and ROS activity of primary and secondary car emissions over eight hours. The different coloured time periods correspond to the driving cycle conditions of the driving cycle. The last hour of each experiment was used as a HEPA blank measurement. No OP and ROS signal was observed for primary exhaust and no gas-phase artefacts were observed.

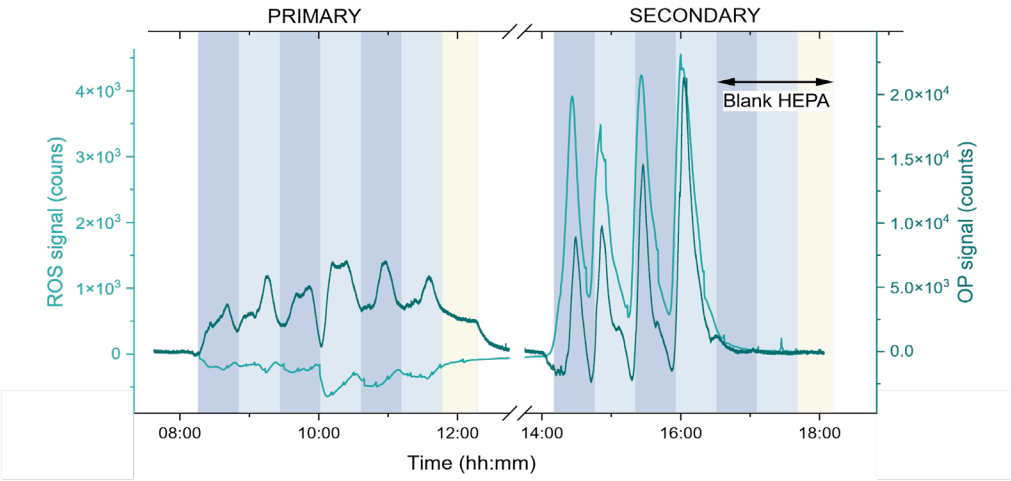

Figure S 5: Blank-corrected signal intensities of OP and ROS activity in primary (left) and secondary (right) RWC emission particles. The colours indicate the addition of a new batch of wood. In total, OP and ROS content were measured in primary and secondary emissions of each six batches of wood. The last batch during primary emissions was left to reach an ember phase (yellow stripes). The last two batches including the ember phase were used as a blank measurement during the secondary emissions.

Activated charcoal was added to the DCFH assay to simulate the high surface area of soot in order to test the hypothesis that the high concentrations in the RWC experiments might cause the HRP enzyme activity to be reduced below blank levels. To get the smallest possible particle size, the charcoal was ground in a mortar. It was then suspended in water and thoroughly mixed to produce a homogenous mixture. For a more controlled environment we used an offline approach as described in Campbell et al., 2021.(Campbell et al., 2021) In order to better align the protocol with the online method of the OPROSI, some adjustments were made. In this revised protocol, soot is added to the HRP and allowed to react for 9 minutes. After this, the mixture is passed through a syringe filter to remove the insoluble particles. Subsequently, DCFH is added and the sample is incubated for an additional 9 minutes. Figure S 2 shows that the addition of charcoal reduced the response of the assay below the blank. This could be an indicator that the negative signals from the OPROSI during primary RWC experiments are caused by the physical presence of insoluble particles in the instrument.

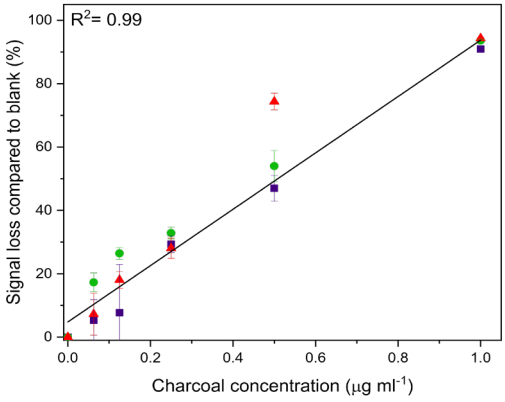

Figure S 6 Soot measurements in triplicates to show the sensitivity loss in %. Error bars are standard deviation of triplicate measurements.

References

Campbell, S. J., Wolfer, K., Utinger, B., Westwood, J., Zhang, Z.-H., Bukowiecki, N., Steimer, S. S., Vu, T. V., Xu, J., Straw, N., Thomson, S., Elzein, A., Sun, Y., Liu, D., Li, L., Fu, P., Lewis, A. C., Harrison, R. M., Bloss, W. J., Loh, M., Miller, M. R., Shi, Z., and Kalberer, M.: Atmospheric conditions and composition that influence PM 2.5 oxidative potential in Beijing, China, Atmos. Chem. Phys, 21, 5549–5573, https://doi.org/10.5194/acp-21-5549-2021, 2021.