CCN-Activation of soot particles after long term exposure to atmospherically relevant ozone concentrations

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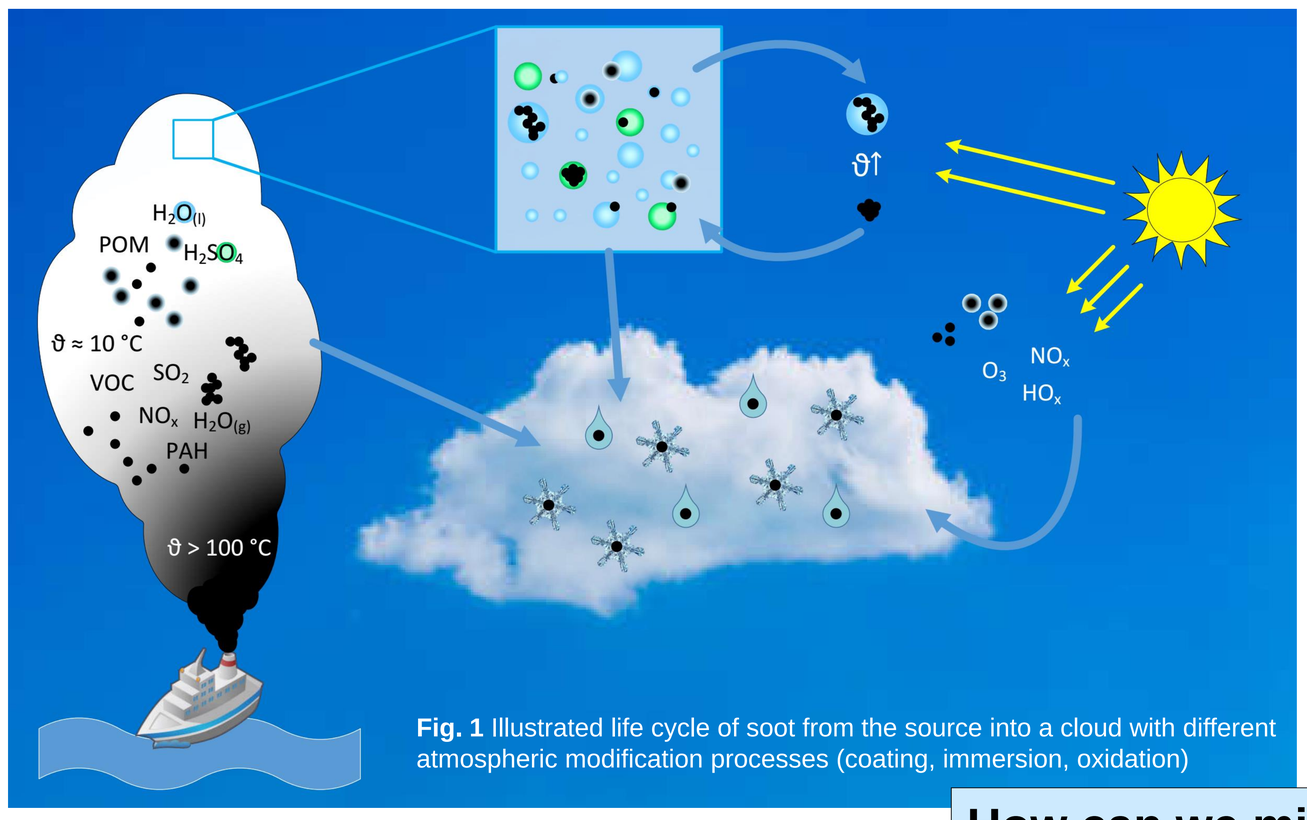


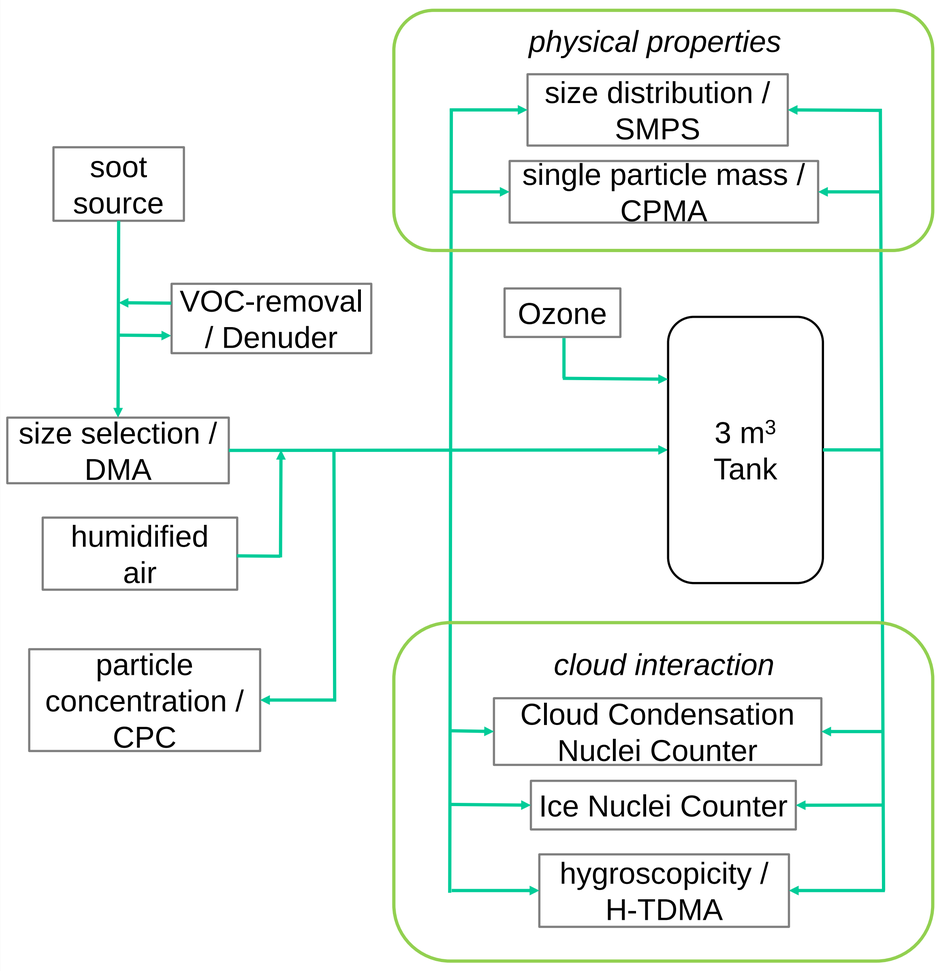
Fig. 1 Illustrated life cycle of soot from the source into a cloud with different atmospheric modification processes (coating, immersion, oxidation)

Introduction

Soot particles that are freshly emitted into the atmosphere are typically hydrophobic and are therefore not able to act as cloud condensation nuclei (CCN). But during an atmospheric lifetime of one week, several processes can occur that change the particle properties significantly. From atmospheric measurement can be concluded that soot particle show some level of CCN-activity. [1]
Aerosol-cloud-interaction contributes significantly to climate change, but also cause the largest uncertainties in climate models. Therefore it becomes obvious that we need a better understanding of the impact of aerosol aging processes.[2]

Here we present a new approach for the investigation of heterogeneous oxidation of aerosols. For that soot particles were exposed to Ozone concentrations of 200 ppb (ambient: 20-120 ppb) for up to 12 hours while the CCN-activity was detected

Setup



How can we mimic several days of atmospheric aging in the lab?

Generally, there are two approaches to achieve/mimic one week atmospheric ageing in the lab. One, the concentration of reactants, e.g. Ozone or OH-radicals can be increased to trigger faster reaction rates. Two, larger aerosol tanks can be constructed to extend the observation time. Both approaches imply their specific challenges. Treating aerosols with high concentrations of oxidants bears the risk that atmospheric processes are not represented well. Extending the observation time by extending the tank volume is often technically and financially limited.
The continuously stirred tank reactor (CSTR) combines long aging times in comparably small chamber volumes, but comes at the cost of a more complex data analysis.

Theory of the CSTR-Setup

The theoretical change of the aerosol concentration while filling and flushing a CSTR and the residence time distribution is fully characterised by the mean residence time τ. The change in the activated fraction (AcFrac) can be obtained from calculating the fraction of Aerosol which is older than the minimum time needed to CCN-activate soot particles (activation time; t_{act}).

While filling the the CSTR with aerosol particles a dynamic equilibrium is reached, which results in a constant AcFrac. When the pre-aged aerosol gets flushed out an exponential increase of the AcFrac is expected.

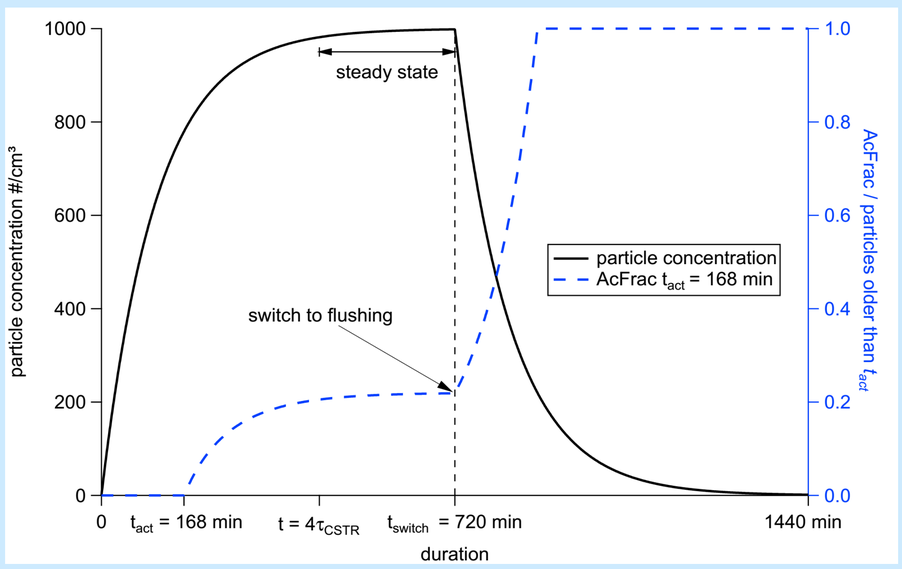
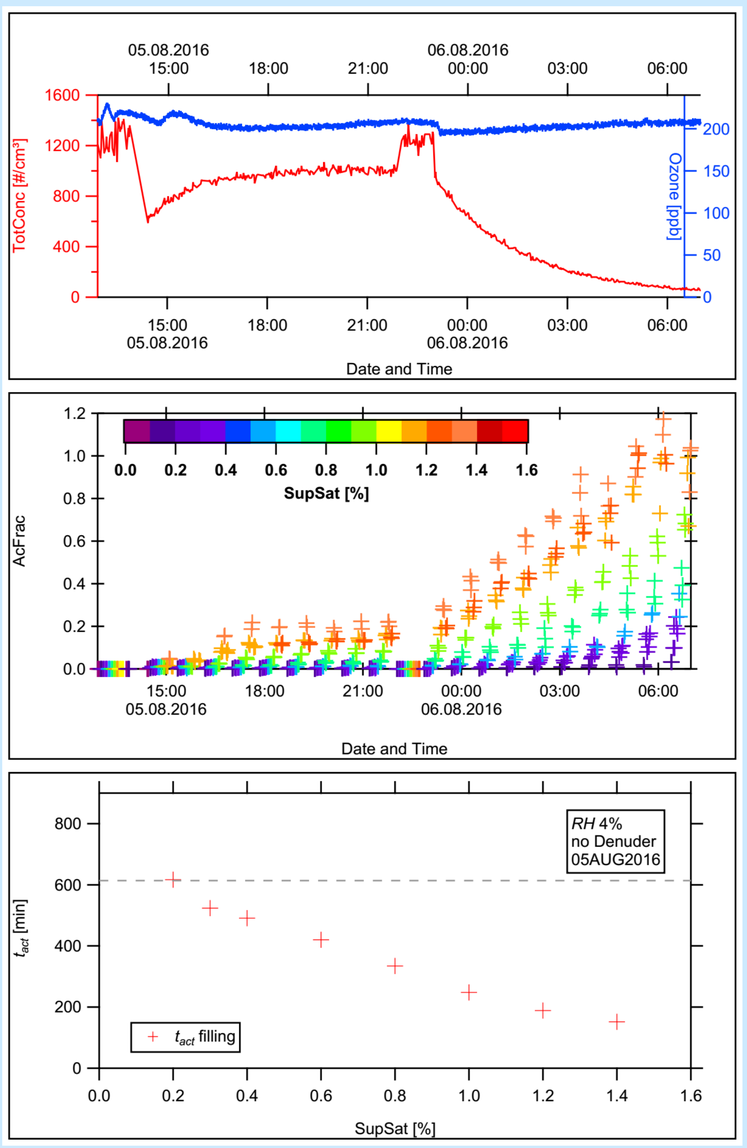


Fig.2 Theoretical change of the particle concentration (black solid) and the AcFrac (blue dashed) for a given t_{act}.



Measurements

The presented CSTR setup was used to expose 100 nm size selected soot particle to Ozone concentrations up to 200 ppb and humidity levels up to 80 % *RH* for exposure times of 12 h. The soot particles were produced with a standardised Propane burner (miniCAST) and were rich in organic carbon. To distinguish between heterogeneous oxidation and condensation of oxidised VOCs onto the particles, the use of a charcoal denuder was optional. In addition to the CCN-activity the aerosol was characterized by the following properties: ice nucleation activity, size distribution, single particle mass, and hygroscopicity.

It was observed that the minimum aging time needed to make a soot particles CCN-active lies in the range of 2 to 10 hours for supersaturations of 1.4 to 0.2 %.

Fig. 3 The upper diagrams show the full raw data set for one day consisting of total aerosol particle concentration (TotConc; red line), Ozone concentration (blue line) and the CCN-AcFrac (color-coded markers) for different supersaturations (SupSat). The elevated particle concentrations before and after filling the CSTR are due to direct measurement of the unaged soot aerosol in a bypass line. The lower diagram shows the obtained t_{act} as a function of the SupSat. The grey dotted line marks the maximum aging time for the particular experiment. All higher t_{act}´s are caused by background noise.

Acknowledgements

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References

[1] IPCC, 2013: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 1535 pp, doi:10.1017/CBO9781107415324.

[2] Hiranuma, N., S. D. Brooks, R. C. Moffet, A. Glen, A. Laskin, M. K. Gilles, P. Liu, A. M. Macdonald, J. W. Strapp, and G. M. McFarquhar (2013), Chemical characterization of individual particles and residuals of cloud droplets and ice crystals collected on board research aircraft in the ISDAC 2008 study, *J. Geophys. Res. Atmos.*, **118**, 6564–6579, doi:10.1002/jgrd.50484.

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