Nanocluster aerosol generation from squalene-ozone reaction

Shen Yang,* Dusan Licina

Human-Oriented Built Environment Lab, School of Architecture, Civil and Environmental Engineering, École Polytechnique Fédérale de Lausanne (EPFL), 1015 Lausanne, Switzerland
shen.yang@epfl.ch

1 INTRODUCTION

Nanocluster aerosols (NCA, particles <3 nm) have strong impacts on climate feedbacks and potentially on human health. Our recent study [1] unveiled the NCA formation owing to gas-phase ozone reaction with human surfaces. However, the underlying mechanisms driving NCA emissions remain unexplored. Squalene is the most abundant compound in human skin lipids that react with ozone, followed by unsaturated fatty acids. This study aims to examine the contribution of squalene-ozone reaction to NCA formation and the influence of ozone and ammonia (NH_3) levels.

2 METHODS

We performed experiments in a 1.9 m^3 stainless-steel climate chamber, which was ventilated by filtered compressed air to ensure low background levels of particles (<100 #/cm^3 below 10 nm) and ozone (<1 ppb). The air temperature was controlled at 24 °C and the relative humidity was maintained at 40%. Ozone was generated by a Jelight 600 UV generator (Jelight Co. Inc., USA). In experiments with NH_3, we injected NH_3 from a gas cylinder (10 ppm, purity >99.9%, Cabagas Inc., CH). Reactants (squalene or C16:1n6) painted on a 0.24 m^2 glass plate were exposed to ozone to investigate NCA formation. A specific quantity of pure squalene (purity >99%, Acros Organics, Thermo Fisher Scientific, USA) or pure C16:1n6 (purity >99%, Cayman, USA) was dissolved in 10 mL methanol and then evenly painted on the glass plate using a glass stick. The painted glass plate was then placed on the stand inside the chamber. Ozone was subsequently injected into the chamber to initiate the reaction for 3 h at 1 h^{-1} air change rate. The reaction was investigated at 15 and 90 ppb ozone to investigate the influence of ozone, whereas NH_3 was injected at 0 and 375 ppb to explore the impact of NH_3. NCA in the size range 1.18-2.81 nm were sampled at 2.5 L/min flow rate and measured in real time at 2-min time interval with a Nano Condensation Nucleus Counter (Airmodus A11 nCNC System, Airmodus, Finland). The ozone concentration inside the chamber was measured with a time resolution of 1 min with an ozone monitor (Model 724, Tanabyte, USA) at 2.0 L/min sampling flowrate. The level of NH_3 was monitored at 30-sec time intervals with a 140 mL/min sampling flowrate using an NH_3 analyzer (LSE NH_3-1700, LSE Monitors, NL).

3 RESULTS AND DISCUSSION

Fig. 1 shows time-series of ozone mixing ratio and NCA size distribution in experiments comparing ozone reaction with squalene and fatty acid C16:1n6. After ozone injection into the chamber, NCA levels in both reaction experiments started to increase. This finding supports the inference from our previous study that ozonolysis of human skin lipid compounds contributes to NCA formation. Although the steady-state ozone levels were similar in both experiments (55 vs. 56 ppb), the steady-state NCA levels were 40 times higher during ozonolysis of squalene relative to C16:1n6, indicating that squalene plays a dominant role in NCA formation when ozone reacts with human skin lipids. The size distributions of NCA further demonstrated

the disparity between the two reactions. Squalene-ozone reaction generated an abundant concentration of NCAs in the smallest size (1.18-1.55 nm), which subsequently grew to larger sizes. In comparison, the ozonolysis of C16:1n6 emitted much lower level of the smallest NCA, with no obvious signals detected for NCAs larger than 1.71 nm. Regarding the influence of ozone and NH_3, results showed that NCA generated by squalene-ozone reaction were positively correlated with ozone level, whereas elevated NH_3 levels were associated with the stronger generation of larger NCA but fewer smallest ones.

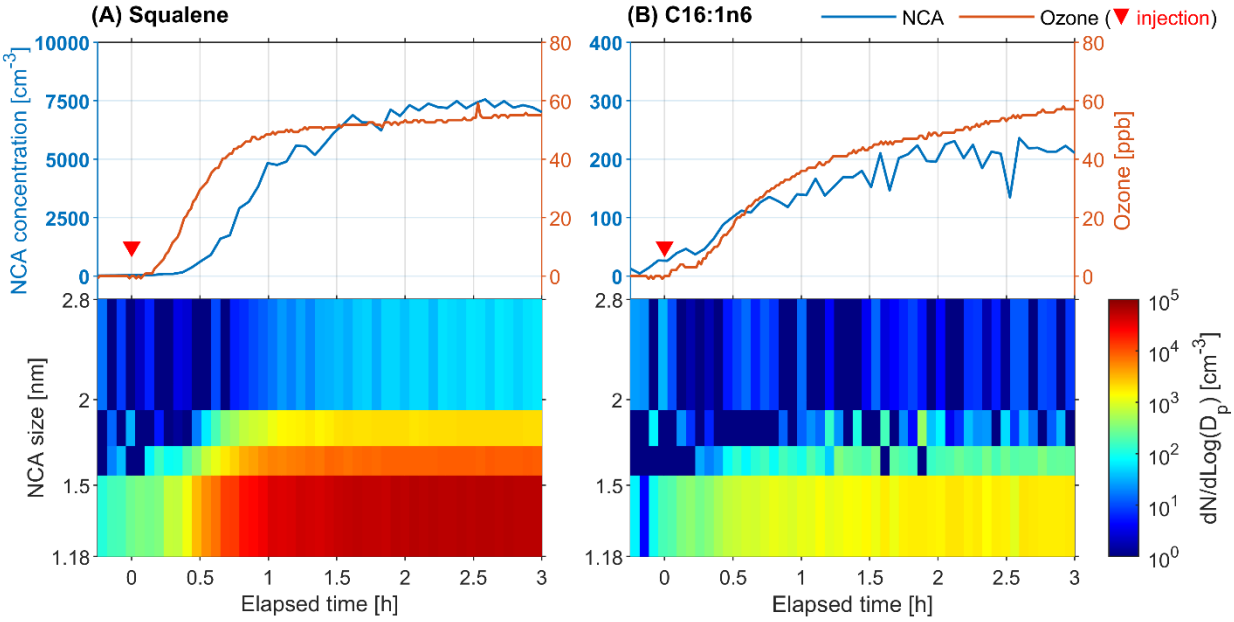

Fig 1. Time-series of total NCA number concentration (1.18-2.8 nm) and ozone (top), and size-resolved NCA concentration (bottom) in experiments comparing ozone reaction with (A) squalene and (B) fatty acid C16:1n6.

4 CONCLUSIONS

The ozonolysis of human skin lipid compounds contributes to NCA formation. With a typical ratio found in human skin lipids (4:1), squalene generated 40 times more NCA than C16:1n6, and thus dominated the NCA formation. NCA generated from squalene-ozone reaction is influenced by ozone and NH_3 levels. This study provides a deeper understanding of the mechanisms driving NCA generation from humans.

ACKNOWLEDGEMENT

The study was funded by the Swiss National Science Foundation (No.: 205321_192086).

5 REFERENCES

[1] Yang S, Licina D, Weschler CJ, Wang N, Zannoni N, Li M, Vanhanen J, Langer S, Wargocki P, Williams J, Bekö G. 2021. *Environmental Science & Technology*. 55(21):14536-45.