Atmospheric Environment 200 (2019) 167–169

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Short communication

Infrequent new particle formation over the remote boreal forest of Siberia

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A B S T R A C T

Aerosol particle number size distributions (PNSD) were investigated to verify, if extremely low-volatility organic vapors (ELVOC) from natural sources alone could induce new particle formation and growth events over the remote boreal forest region of Siberia, hundreds of kilometers away from significant anthropogenic sources. We re-evaluated observations determined at a height of 300 m of the remote observatory ZOTTO (Zotino Tall Tower Observatory, http://www.zottoproject.org). We found that new particle formation events occurred only on 11 days in a 3-year period, suggesting that homogeneous nucleation with a subsequent condensational growth could not be the major process, maintaining the particle number concentration in the planetary boundary layer of the remote boreal forest area of Siberia.

1. Investigation

In the study by Ehn et al. (2014), the formation of low-volatile secondary organic aerosol from biogenic precursors was described. The authors studied the oxidation process of several monoterpenes, such as α-pinene, and other VOCs (volatile organic carbon) under atmospherically relevant conditions in the "Jülich Plant Atmosphere Chamber" (JPAC). This work was a milestone for a better understanding of atmospheric oxidation processes of VOC as aerosol particle precursors, especially over boreal forest regions with negligible anthropogenic influence. A direct pathway of several biogenic VOCs (monoterpenes) was found for the formation of extremely low-volatility organic vapors (ELVOC). The major outcome of this study was to quantify more accurately the effects of changes in biogenic VOC emission, specifically on new particle formation and the abundance of cloud condensation nuclei. The authors propose that the ELVOCs formed from monoterpene oxidation will help to explain the appearance and the subsequent rapid growth of new particles observed in forested regions. Such new particle formation events have been frequently observed at forest research station Hyytiälä in Finland in the past (Kulmala et al., 2012). However, Hyytiälä might not be a fully representative observational site to prove new particle formation induced by ELVOCs alone, since it might be still influenced by gaseous sulfuric acid from anthropogenic SO_2. It is probably hard to prove how much anthropogenic influence is needed to initiate new particle formation over a pristine boreal forest.

The objective of our investigation was to verify whether the above-mentioned assumptions and conclusions could be confirmed by observations at an even more remotely located boreal forest location. For this purpose, we re-analyzed particle number size distribution (PNSD) measurements from the remote Zotino Tall Tower Observatory (ZOTTO), located in the Siberian forest at 60.8° N; 89.35° E. The underlying data set was described in Heintzenberg et al. (2011), but had previously not been analyzed with respect to the phenomenon of the particle formation process. At ZOTTO, sampling of aerosol particles takes place at the top of a tall meteorological tower at 300 m height so that measurements are representative for a considerable area within the Siberian ecosystem. Anthropogenic sources are relatively scarce and far from each other within a range of a few hundred kilometers around ZOTTO (Heintzenberg et al., 2011). The ZOTTO measurements are more representative for new particle formation in the boundary layer and for a larger region compared to measurements taken directly on ground.

The PNSDs were measured using a TROPOS-type mobility particle size spectrometer as described in Wiedensohler et al. (2012). Losses of ultrafine particles due to diffusion in the 300 m long sampling tube can be corrected down to 10 nm, according to Birmili et al. (2007). This means that ZOTTO PNSDs can be analyzed at least qualitatively for new

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https://doi.org/10.1016/j.atmosenv.2018.12.013
Received 4 July 2018; Received in revised form 9 December 2018; Accepted 11 December 2018
Available online 19 December 2018

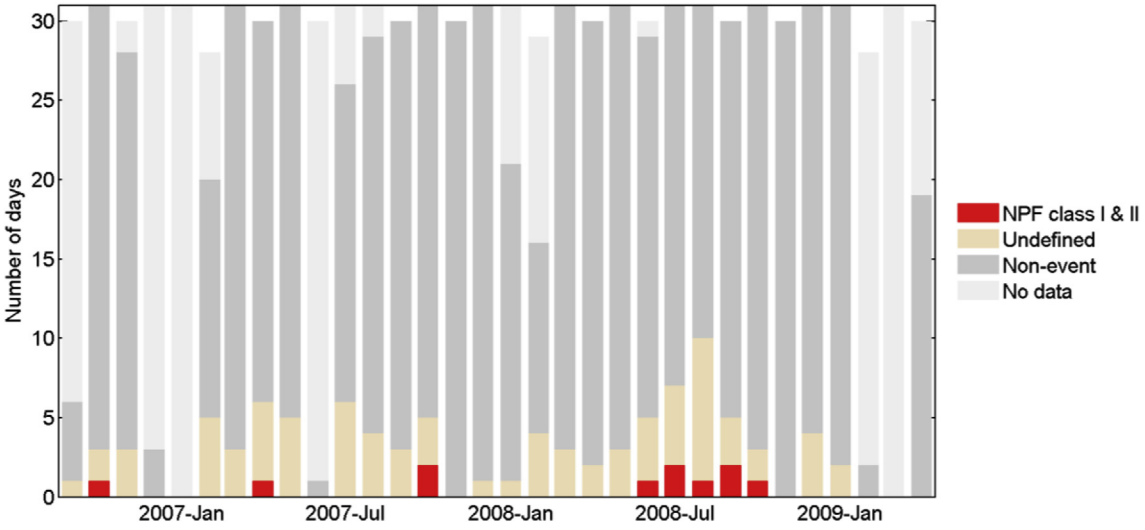

Fig. 1. Statistics of new particle formation (NPF) events in each month during the experiment period.

particle formation events with a subsequent growth down to a particle size of 15 nm. Chi et al. (2013) provided an analysis of particle size distributions under pristine and polluted conditions at ZOTTO. However, an analysis about new particle formation was not done in detail yet.

We thus re-analyzed all PNSDs from Sep. 2006 to Apr. 2009 using the method described by Dal Maso et al. (2005). This method classifies all days with complete data coverage into new particle formation events with a subsequent growth as well as undefined and non-event cases. Fig. 1 shows a time series describing the total number of days per month according to this classification. Days with new particle formation are marked in red. Out of 752 days with valid measurements, only 11 were new particle formation events, 83 were undefined and 658 were non-event days. Out of the 11 days with a new particle formation event, formation rates and growth rates could be calculated only for six event days. The other five event days showed a too weak new particle formation or had strong fluctuations in particle number concentration. The average formation rate (J15) and growth rate of the six days with clear events are $2.30*10^{-2}$ $1/cm^3s^1$ (max. $6.36*10^{-2}$ and min. $5.20*10^{-3}$) and 2.44 nm/h^1 (max. 4.59 nm/h^1 and min. 0.79 nm/h^1), respectively. The 2-h average condensational sink before the nucleation of the six events is $1.98*10^{-3}$ s^{-1} (max. $5.40*10^{-3}$ s^{-1} and min. $2.12*10^{-4}$ s^{-1}).

Regionally relevant new particle formation with a significant subsequent particle growth appeared only in 1.5% of all valid measurement days. These statistics are in contrast to the results obtained in Hyytiälä, Finland, where new particle formation events were much more frequent. It also suggests that new particle formation events do not seem to be a major factor maintaining the particle number concentration in the planetary boundary layer of the remote forested area of Siberia.

If one would add the “undefined nucleation events”, which are either classified a) for a period of nucleation mode particles, which did not grow, or b) when there has been a particle growth, starting in the Aitken mode size range. For these type events, no banana-shaped particle nucleation & growth were visible, which would be representative for a regional new particle formation event. In the most optimistic case there might then be an upper limit of 10% probability for a new particle formation event in the Central Siberian forest. For Hyytiälä, there have been done few studies to determine the nucleation frequency. In the article by Dal Maso et al. (2005), an average frequency of 24% for particle formation day was determined, which is significantly higher than at ZOTTO in central Siberia. Nieminen et la. (2014) stated: “During the years 1996–2012, the total number of NPF event days was 1418. Annually, the number of event days varied in the range 60–120. The highest number of NPF events was observed in the years 2002–2004 when during each year more than 110 days were categorized as event days. The NPF events were least frequent in 1998 and 2010 with less than 70 NPF event days in both years. The average yearly fraction of NPF event days was 23%.” They additionally concluded that “the probability of the NPF was at the maximum of about 40%–50% during spring (March, April and May), and a second maximum of about 30%–40% took place in September. In winter, < 10% of the days were NPF event days.”

The ZOTTO result of infrequent new particle formation are supported by the mean diurnal variation of the PNSD of the four seasons. The contour plots in Fig. 2 show for all seasons the same structure of the PNSD with a peak diameter around 100 nm during the course of the day. This suggests that the aerosol particles are aged and transported over a long distance. However, during the warmer periods, the particle number concentration is enhanced. Photo-chemically induced new particle formation took possibly place elsewhere with growth during the long-range transport.

We have shown that in the remote boreal forest of Siberia area with supposedly abundant monoterpenes, but negligible anthropogenic influence, new particle formation events were only infrequently observed. This result suggests that the oxidation of monoterpenes proposed by Ehn et al. (2014) might be not the major contributor of new atmospheric particles over this remote forest area. This reason for this could be either a) that the reaction condition or oxidants required by the reaction pathway cannot be easily achieved in the real atmosphere, or b) that particle formation and growth over remote forested areas are more complex and cannot be simply described by the formation of ELVOCs. Given that organic matter makes up the largest fraction of the aerosol particle composition over the Siberian region during the growing season (Mikhailov et al., 2015), it is, however, likely that BVOC oxidation contributes to the growth of pre-existing particles, similar to what has been shown for the Amazon rainforest (Pöschl et al., 2010).

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

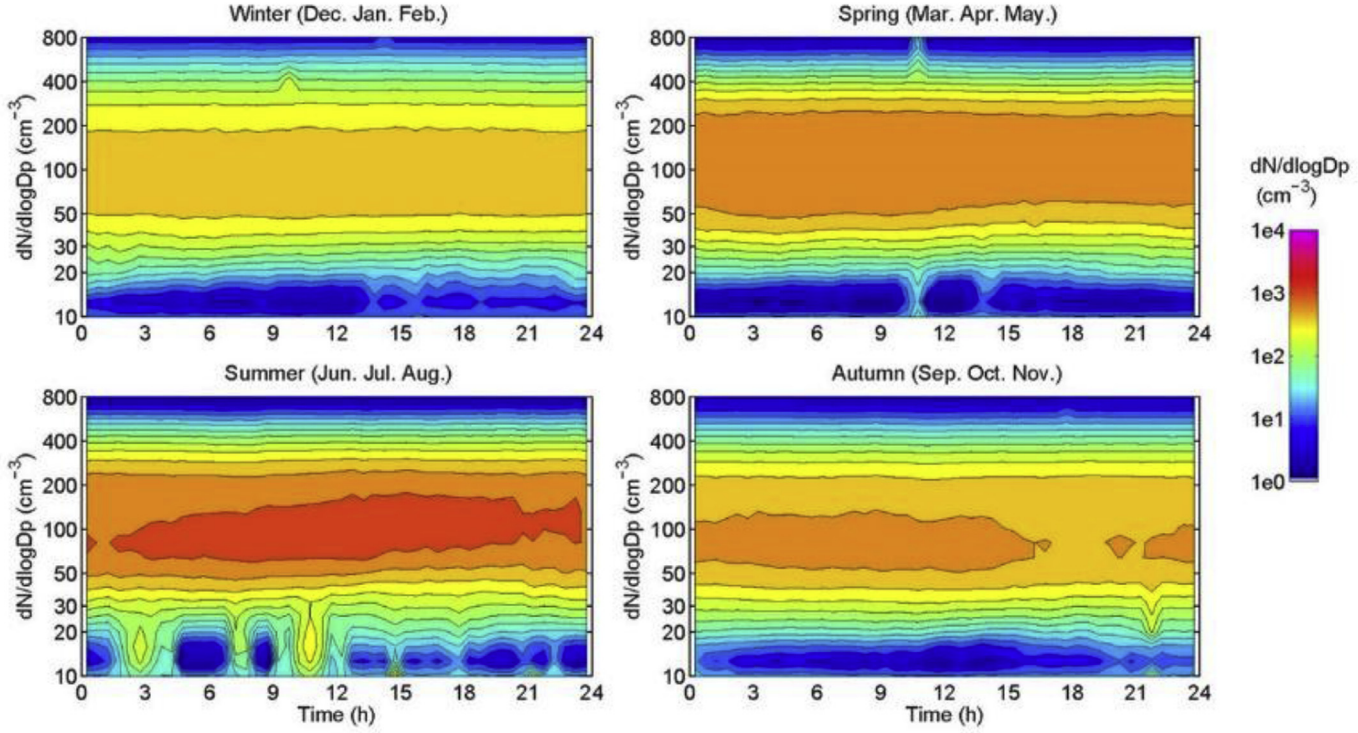

Fig. 2. Average diurnal variation of PNSDs at 300 m for the four seasons.

Ackowledgements

The ZOTTO project was funded by the Max Plank Society through the International Science and Technology Center (ISTC) partner project #2757p within the framework of the proposal 'Observing and Understanding Biogeochemical Responses to Rapid Climate Changes in Eurasia. The aerosol measurements have been additionally funded by the "Deutsche Forschungsgemeinschaft" under the title "Climatically Relevant Atmospheric Aerosols over Siberia" und the project number 59451000.

References

Birmili, W., Stopfkuchen, K., Hermann, M., Wiedensohler, A., Heintzenberg, J., 2007. Particle transport losses in a 300 m inlet pipe for sampling atmospheric aerosols from a tall meteorological tower. Aerosol Sci. Technol. 41, 811–817.

Chi, X., Winderlich, J., Mayer, J.C., Panov, A.V., Heimann, M., Birmili, W., Heintzenberg, J., Cheng, Y., Andreae, M.O., 2013. Long-term measurements of aerosol and carbon monoxide at the ZOTTO tall tower to characterize polluted and pristine air in the Siberian taiga. Atmos. Chem. Phys. 13 (24), 12271–12298.

Dal Maso, M., Kulmala, M., Riipinen, I., Wagner, R., Hussein, T., Aalto, P.P., Lehtinen, K.E.J., 2005. Formation and growth of fresh atmospheric aerosols: eight years of aerosol size distribution data from SMEAR II, Hyytiala, Finland. Boreal Environ. Res. 10 (5), 323–336.

Ehn, M., Thornton, J.A., Kleist, E., Sipila, M., Junninen, H., Pullinen, I., Springer, M., Rubach, F., Tillmann, R., Lee, B., Lopez-Hilfiker, F., Andres, S., Acir, I.H., Rissanen, M., Jokinen, T., Schobesberger, S., Kangasluoma, J., Kontkanen, J., Nieminen, T., Kurten, T., Nielsen, L.B., Jorgensen, S., Kjaergaard, H.G., Canagaratna, M., Dal Maso, M., Berndt, T., Petaja, T., Wahner, A., Kerminen, V.M., Kulmala, M., Worsnop, D.R., Wildt, J., Mentel, T.F., 2014. A large source of low-volatility secondary organic aerosol. Nature 506 (7489), 476–479.

Heintzenberg, J., Birmili, W., Otto, R., Andreae, M.O., Mayer, J.C., Chi, X., Panov, A., 2011. Aerosol particle number size distributions and particulate light absorption at the ZOTTO tall tower (Siberia), 2006-2009. Atmos. Chem. Phys. 11 (16), 8703–8719.

Kulmala, M., Petaja, T., Nieminen, T., Sipila, M., Manninen, H.E., Lehtipalo, K., Dal Maso, M., Aalto, P.P., Junninen, H., Paasonen, P., Riipinen, I., Lehtinen, K.E.J., Laaksonen, A., Kerminen, V.M., 2012. Measurement of the nucleation of atmospheric aerosol particles. Nat. Protoc. 7 (9), 1651–1667.

Mikhailov, E.F., Mironov, G.N., Pohlker, C., Chi, X., Kruger, M.L., Shiraiwa, M., Forster, J.D., Poschl, U., Vlasenko, S.S., Ryshkevich, T.I., Weigand, M., Kilcoyne, A.L.D., Andreae, M.O., 2015. Chemical composition, microstructure, and hygroscopic properties of aerosol particles at the Zotino Tall Tower Observatory (ZOTTO), Siberia, during a summer campaign. Atmos. Chem. Phys. 15 (15), 8847–8869.

Nieminen, T., Asmi, A., Dal Maso, M., Aalto, P.P., Keronen, P., Petaja, T., Kulmala, M., Kerminen, V.M., 2014. Trends in atmospheric new-particle formation: 16 years of observations in a boreal-forest environment. Boreal Environ. Res. 19, 191–214.

Pöschl, U., Martin, S.T., Sinha, B., Chen, Q., Gunthe, S.S., Huffman, J.A., Borrmann, S., Farmer, D.K., Garland, R.M., Helas, G., Jimenez, J.L., King, S.M., Manzi, A., Mikhailov, E., Pauliquevis, T., Petters, M.D., Prenni, A.J., Roldin, P., Rose, D., Schneider, J., Su, H., Zorn, S.R., Artaxo, P., Andreae, M.O., 2010. Rainforest aerosols as biogenic nuclei of clouds and precipitation in the Amazon. Science 329 (5998), 1513–1516.

Wiedensohler, A., Birmili, W., Nowak, A., Sonntag, A., Weinhold, K., Merkel, M., Wehner, B., Tuch, T., Pfeifer, S., Fiebig, M., Fjäraa, A.M., Asmi, E., Sellegri, K., Venzac, H., Villani, P., Laj, P., Aalto, P., Ogren, J.A., Swietlicki, E., Roldin, P., Williams, P., Quincey, P., Hüglin, C., Fierz-Schmidhauser, R., Gysel, M., Weingartner, E., Riccobono, F., Santos, S., Grüning, C., Faloon, K., Beddows, D., Harrison, R., Monahan, C., Jennings, S.G., O'Dowd, C.D., Marioni, A., Horn, H.-G., Keck, L., Jiang, J., Scheckman, J., McMurry, P.H., Deng, Z., Zhao, C.S., Moerman, M., Henzing, B., Leeuw, G. d., Löschau, G., Bastian, S., 2012. Mobility particle size spectrometers: harmonization of technical standards and data structure to facilitate high quality long-term observations of atmospheric particle number size distributions. Atmospheric Measurement Techniques 5, 657–685.