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**Nucleation parameter
indicates future rise**

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Ambient new particle formation parameter indicates potential rise in future events

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Abstract

Atmospheric new particle formation is a general phenomenon observed over coniferous forests. So far nucleation is described as a function of gaseous sulfuric acid concentration only, which is unable to explain the observed seasonality of nucleation events at different measurement sites. Here we introduce a new nucleation parameter including ozone and water vapor concentrations as well as UV-B radiation as a proxy for OH radical formation. Applying this new parameter to field studies conducted at Finnish and German measurement sites it is found capable to predict the occurrence of nucleation events and their seasonal and annual variation indicating a significant role of organics. Extrapolation to possible future conditions of ozone, water vapor and organic concentrations leads to a significant potential increase in nucleation event number.

1 Introduction

Atmospheric new particle formation is a common phenomenon occurring anywhere close to the Earth's surface as well as above (Kulmala et al., 2004a). These new particles are expected to originate from a variety of different sources and gaseous precursors such as sulfuric acid, reactive volatile organic compounds (VOCs) and iodine oxides. However the current limitation for direct chemical analysis of the tiniest aerosol particles and clusters is at around 10 nm in diameter. The requested mass for analysis makes it necessary to use correlation methods between potential gaseous precursors and apparent number concentration of the smallest particles. A good correlation is assumed to be an indicator for the reliability of a postulated nucleation mechanism but not a sufficient criterion, because of the similar behavior of different gases.

Once new clusters and particles are formed they may grow to sizes, at which they will affect cloud properties and the radiation budget at the Earth's surface. The corresponding effects are some of the key uncertainties in correctly describing climate

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changes and are expected to be 2–14 W m⁻² for boreal forest sites including cloud effects (Kurtén et al., 2003; Spracklen et al., 2008). Most of these effects caused by the grown aerosols are independent of the initial nucleation process and gases involved because the growth is predominantly caused by other gases. Nevertheless, the amount of newly formed particles that can act as cloud condensation or ice nuclei depends on the nucleation mechanism and the involved trace gases responsible for the further growth.

So far ambient nucleation is commonly treated as a function of sulfuric acid concentration either linearly or squared (Kulmala et al., 2006; Sihto et al., 2006; Riipinen et al., 2007):

$$J = A \cdot [\text{H}_2\text{SO}_4] \quad (1)$$

$$J = K \cdot [\text{H}_2\text{SO}_4]^2 \quad (2)$$

Since sulfuric acid is a gas of low volatility, thus of short atmospheric lifetimes and is formed from the reaction of predominantly sulfur dioxide and OH, it might serve here as a marker for any compound that is formed dependent on radiation intensity and reaction of OH with a precursor gas. The exact nature of this gas remains speculative. The correlation coefficients *A* and *K* obtained from field studies vary by orders of magnitudes between different sites and at different times throughout the year for instance for boreal forests, which cover about 15% of the global land surface. Bonn et al. (2008) explained this variation for the Finnish boreal forest site in Hyytiälä (SMEAR II Kulmala et al., 2001; Hari and Kulmala, 2005) by a nucleation caused by the reaction of biogenic VOCs (sesquiterpenes) with ozone. This is in line with the observations made by Went (1960) half a century earlier explaining the blue haze phenomenon in the United States. In addition to the study of Went (1960) Bonn and Moortgat (2003) included a suppressing effect of water vapor on nucleation, i.e. the more water vapor (higher humidity) the less nucleation caused by a nucleation precursor the so-called stabilized Criegee Intermediate (sCI Criegee, 1975). Bonn et al. (2008) merged these laboratory

findings with the observations reading sulfuric acid (Sihto et al., 2006; Riipinen et al., 2007) leading to a postulated nucleation mechanism displayed in Fig. 1. If this occurs as postulated one would expect a local minimum during the most humid period during summer, in which VOCs are emitted strongest, and two maxima during the transition periods in spring and autumn.

In this study we formulate a new nucleation parameter using proxies for organic (related to organic-ozone reactions) and OH radical contribution to atmospheric particle formation in order to investigate its influence on the observed seasonality and annual behavior of these events. Finally we use this parameter to investigate the possible future evolution of new particle formation in and over boreal and coniferous forests.

2 Nucleation parameter

If nucleation is initiated by the reaction of a reactive VOC such as a terpene and ozone as it is known from Went (1960), the nucleation rate depends on terpene and ozone concentrations and on their reaction rate $k_{\text{terpene}+\text{ozone}}$. As shown in Fig. 1 the critical intermediate compound is the stabilized Criegee Intermediate (sCI), a biradical, which predominantly reacts with water vapor ($k_{\text{sCI}+\text{water}}$) in the atmosphere resulting in a lifetime of sCIs of about one second. A steady-state approach of source equals sink leads to the following equation:

$$[\text{sCI}] = \frac{k_{\text{terpene}+\text{ozone}} \cdot [\text{O}_3][\text{terpene}] \cdot \text{stab.fraction}}{k_{\text{sCI}+\text{H}_2\text{O}} \cdot [\text{H}_2\text{O}]} \quad (3)$$

If we divide this by the terpene concentration, which is the one most difficult to observe, we get the ratio of ambient sCI to terpene. This ratio appears in the nucleation description by Bonn et al. (2008) and is defined as the organic nucleation parameter A (organic NP A):

$$\text{organic NPA} = \frac{[\text{sCI}]}{[\text{terpene}]} \quad (4)$$

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$$= \frac{k_{\text{terpene+ozone}} \cdot [\text{O}_3] \cdot \text{stab.fraction}}{k_{\text{sCl+H}_2\text{O}} \cdot [\text{H}_2\text{O}]} \quad (5)$$

It essentially depends on ozone and water vapor concentrations, which are easy to observe. Additionally terpenes are needed, which can be represented by their expected emission in a second organic parameter B :

$$5 \text{ organic NPB} = \frac{k_{\text{terpene+ozone}} \cdot [\text{O}_3] \cdot \text{stab.fraction}}{k_{\text{sCl+H}_2\text{O}} \cdot [\text{H}_2\text{O}]} \cdot \exp(\beta \cdot (T - 30^\circ\text{C})) \quad (6)$$

Since numerous studies have revealed the dependence of nucleation on hydroxyl radical (OH) derived compounds, we define the complete nucleation parameters NP A and NP B by the product of the organic part (Eq. 5 and 6) and of the ultraviolet B radiation flux, necessary for OH production.

$$\text{NPA} = \text{organic NPA} \cdot \text{UV B}$$

$$= \frac{k_{\text{terpene+ozone}} \cdot [\text{O}_3] \cdot \text{stab.fraction}}{k_{\text{sCl+H}_2\text{O}} \cdot [\text{H}_2\text{O}]} \cdot \text{UV B} \quad (7)$$

$$10 \text{ NPB} = \text{organic NPB} \cdot \text{UV B}$$

$$= \frac{k_{\text{terpene+ozone}} \cdot [\text{O}_3] \cdot \text{stab.fraction}}{k_{\text{sCl+H}_2\text{O}} \cdot [\text{H}_2\text{O}]} \cdot \exp(\beta \cdot (T - 30^\circ\text{C})) \cdot \text{UV B}$$

Because UV B measurements are not always available, we exemplarily use 1% of the global radiation as a proxy for UV B in situations, where UV B data are missing and discuss the effect.

15 The emission of terpenes used in Eqs. (6) and (7) has been postulated by Guenther et al. (1995) and Tarvainen et al. (2005) to be exponentially dependent on temperature. However, the exact coefficient is not well known for instance in the case of sesquiterpenes. Therefore we consider always two nucleation parameters with A) terpenes to be present in sufficient amounts if the temperature is higher than the freezing point and B) terpene emission in accordance to Guenther et al. (1995). Since

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terpenes are of various different types and possess a different reactivity we focused on the one most likely to cause ambient nucleation, i.e. the sesquiterpene ($C_{15}H_{24}$) β -caryophyllene. For other environments the rate constant and stabilization fraction can be exchanged according to the predominant terpene. For β -caryophyllene we use the following values: $k_{\text{terpene}+\text{ozone}}=1.16\times 10^{-14}\text{ cm}^3\text{ molecule}^{-1}\text{ s}^{-1}$ (Shu and Atkinson, 1994), stabilised fraction = 0.94 (approximated from Chuong et al., 2004) as well as $k_{\text{SCL}+\text{water}}=10^{-17}\text{ cm}^3\text{ molecule}^{-1}\text{ s}^{-1}$ (Bonn et al., 2008). It should be noted that this parameter investigates the very first steps of particle formation but not the subsequent growth (Fig. 1) one can use the parameters for the smallest particles only, not for larger ones.

3 Intercomparison with field measurements

In order to evaluate the performance of the NP we intercompare the calculations with representative measurements for different environments: (a) SMEAR II station, Hyytiälä (Finland) (Hari and Kulmala, 2005) and (b) Taunus observatory (Mt. Kleiner Feldberg, 812 m a.s.l., Germany). Both stations are representative sites for remote locations, with the Finnish station to be very remote and the German one facing more anthropogenic affected air masses. Aerosol particle size distribution as well as meteorological parameters and trace gases are monitored continuously by the Hessian Institute for Environment and Geology (HLUG, see supporting online information). Since the German aerosol measurement period is much less extensive than the Finnish one, we include the German station for short term intercomparison only and the Finnish station additionally for longer periods.

3.1 Daily intercomparison

Aerosol size distribution measurements at Taunus observatory are monitored since the end of February this year, while measurements in Finland started in January 1996 with the most extensive dataset of any boreal forest site.

5 For the Taunus observatory data we find a generally good agreement between a rise in both NP and in total particle number concentration (Fig. 2, top), which is dominated by secondary sources at the hill site and which is most effective at winds from remote regions in contrast to the Frankfurt area on the opposite site. Correlations are found independent of the time of the year, in March as well as in July. Since direct UV B
10 measurements are not available, 1% of the obtained global radiation is used instead, which is a potential source of uncertainty for the calculated values. For intercomparison the organic NP *A* and *B* (see Eqs. 5 and 6) are shown as well. Please note that the organic NP *A* displays sometimes a shift in time to the nucleation parameter, caused by the remarkable impact of the daily cycle of solar radiation. If this is apparent, two
15 particle number concentration maxima correlating with the maxima of the individual parameters are visible. This indicates to possible particle formation mechanisms with one more related to UV radiation and the other to biogenic hydrocarbon oxidation.

A similar but clearer behavior is found for Hyytiälä in Finland, for which UV B radiation data are available (Fig. 2, bottom). It is obvious that the parameter NP *A* (Eq. 7) serves
20 as an upper limit with the parameter NP *B* (Eq. 7) much closer to the pattern of the particle number concentration, indicating a notable contribution of biogenic terpenes to the formation process. The difference in the magnitude of the nucleation parameter is likely to be caused by different terpene emissions at both sites. Using the organic part of NP *A* only, there is a clear change between necessary values for nucleation start
25 during winter or early spring and summer with a drop in necessary NP *A* by a factor of two to three during the middle of the year. Nevertheless, the increase in nucleation probability is compensated by the suppressing effect of water vapor, increasing towards the warmer season.

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3.2 Seasonal scale

For the seasonal behavior we use the dataset from 1996–2007 at Hyytiälä (partially in Dal Maso et al., 2005). Figure 3 presents the event frequency observed at the site and the organic individual NPs. A good agreement of the organic NP A at the first maximum during springtime is obvious, while the second in August is missing. The March maximum is caused by high ozone and low water vapor concentrations, both strongly supporting nucleation. The mismatch during August might be caused by a) the neglect of the organic hydrocarbon contribution, which is supposed to be maximum during the warmest period (June–August) and b) by the suppressing effect of further trace gases such as organic acids on nucleation. Regarding a) one should be careful applying the excellent Guenther algorithm for monoterpenes (Guenther et al., 1995) gained at notably higher temperatures to boreal environmental conditions. If any decomposition of organic material or water stress effects have an additional significant contribution, the seasonal relationship would be miscalculated completely. However, with respect to annual behavior the temperature effect the stress dependency (reference to supporting online material: <http://www.atmos-chem-phys-discuss.net/9/673/2009/acpd-9-673-2009-supplement.pdf>) of the hydrocarbon emission becomes more important. By increasing the emission mainly during autumn the otherwise suppressed nucleation can take place.

Regarding b): As shown in Fig. 1 formaldehyde is one of the major compounds leading to nucleation. The ratio of formaldehyde to organic acids calculated for Hyytiälä indicates two maxima (Dal Maso et al., 2005) in spring and in autumn (see Supplement: <http://www.atmos-chem-phys-discuss.net/9/673/2009/acpd-9-673-2009-supplement.pdf>), while during June and July the organic acids and water vapor counteract the formation of the nucleation core leading to a reduction in event frequency. This is not included within the parameter given, but can be done, if measurements are available. The other unknown aspect is the exact emission dependency on temperature of the corresponding hydrocarbon and the resulting mixing

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within the boundary layer. Using different temperature coefficients (Guenther et al., 1995; Tarvainen et al., 2005) shifts the maximum found in March towards the summer. The organic NP B and both parameters correlated with UV radiation B as a proxy for OH do not explain the general pattern. This is most likely caused by the dominant effect of water vapor on nucleation and the availability of OH during daytime, because it is not the strength, which is investigated, but the occurrence of new particle formation.

3.3 Annual scale

A further important point worth consideration is the annual time scale. If the parameter is able to predict changes within a changing environment it should be able to explain changes between different years as found at Hyytiälä (Dal Maso et al., 2005).

In order to investigate this we calculate the mean NPs of the most intense nucleation periods – spring (March to May) and autumn (July to September) – for the SMEAR II site for the years from 1996 until 2007 and intercompare these with the number of nucleation event days found. The results are shown in Fig. 3 at the bottom. The pattern is nicely reproduced by the new organic NP B (including the terpene emission), while the other parameters show a tendency with a huge scattering but no significant trend. A deviation is visible for 2006, during which the NP increased substantially from summer on with clear maximum peaks. However, the intensity of nucleation is not investigated in this plot but the occurrence. This can be explained best with two situations: (i) a high peak in a parameter during a short time affecting the mean value and (ii) a significantly flatter peak on a different day. Imagine nucleation to occur on both days independent if a critical value is exceeded by 10 or by 100%. Hence strong variation with sharp peaks complicate the interpretation.

The good match of NP B is caused by an elevated temperature as well as ozone and water vapor mixing ratios. A small deviation can be seen for the years with gaps in data of especially UV B – 1997 and 2002. Since these gaps were exactly during the periods of highest nucleation intensity the mean parameter calculated without the values for the missing period can notably altered.

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4 Future predictions

Finally we apply the postulated NPs to estimate the future evolution of nucleation event day numbers affected by climate change. Is this common phenomenon fading out or gaining in intensity?

5 In order to calculate a potential change in nucleation event number, the changes in ozone, organics, water vapor and radiation need to be considered. The fourth assessment report of the intergovernmental panel on climate change (IPCC report, 2007) expects a future ascent of the average surface temperature by about +3 K, with higher values towards the poles (boreal regions) and smaller towards the equator. Similarly
10 a rise in tropospheric ozone is expected to occur as was observed by different studies such as Jaffe et al. (2003) with an increase of 1.4% per year. The exact behavior of ozone in a changing environment depends on the NO_x and VOC emissions and the fact, if it is a NO_x or VOC limited ozone production. This varies notably over the area of interest. However, in boreal regions and coniferous forest areas there is a tendency
15 to VOC limited ozone production, which causes the changes in VOC emission to be reflected in the change in ozone production.

For a detailed investigation of this aspect we use two sites: (i) the Taunus observatory and (ii) the SMEAR II station in Hyytiälä (Southern Finland). At the Taunus observatory ozone and relative humidity were monitored continuously by the Hessian
20 Institute for Environment and Geology (HLUG) since 1996. This station is expected to be representative for a medium remote location of a coniferous forest in the mid-latitudes, at which the nucleation is expected to occur. From the data analysis of ozone and temperature data we find a cubic correlation, since ozone production is driven by the emission of VOCs, which is predominantly a result of temperature stress on plants.
25 This is available in the supporting online information (see supporting online information). For instance an increase of 4.2 ppbv K⁻¹ is observed at 20°C, which represents an increase of 6%, which is about twice as much as found for Hyytiälä conditions. At the same time water vapor volume mixing ratio rises between 6 and 7.5% per Kelvin at

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a similar relative humidity at warmer temperatures according to the Goff-Gratch formula (Pruppacher and Klett, 1997).

From the current knowledge about terpene emissions (Guenther et al., 1995; Tarvainen et al., 2005) it is known that the emission of the most important terpene class, i.e. the monoterpenes can be described by an exponential temperature dependency of $\exp(\beta \cdot (T - 30^\circ\text{C}))$ with $\beta = 0.09 \text{ K}^{-1}$. Thus the emission increases by approximately 9–10% per Kelvin. Sesquiterpenes are proposed to have a twice as large temperature dependency (Tarvainen et al., 2005). The UV B radiation strength is a function of latitude and time of the year and of the distance between Earth and sun. The latter appears like a weak sinusoidal pattern with a 12 years cycle. But no significant changes are to be expected regarding this within the next century. Only an intensified cloud formation and extended cloud lifetime would cause a reduction. Therefore, we omit any UV B changes in the following considerations.

We apply the dependence of monoterpenes as a conservative estimate and expect the minimum relative change in the nucleation parameters as displayed in Fig. 4. In the upper graph the change in NP B is shown for different reference temperatures, thus if one compares a change by for instance 2 K at 0 or 20°C. From this plot we expect the climate warming effect on NP B – organic as well as including UV B radiation – to be a relative increase between 4 and 9% at a temperature change of +1 K. The effect is highest for the largest temperatures because of the increase of terpene emissions with increasing temperature, which is higher than the one of ozone and water vapour, which nearly cancel out. However this is highly dependent on the local increase in ozone, in terpene emissions and water vapour concentrations. These need to be understood well to make severe predictions for any site of interest.

The individual contribution of ozone, temperature and water vapour to the change in NP is investigated in the lower part of Fig. 4. Therein, NP relative changes are displayed for a reference temperature of 20°C, (i) as shown in the top plot for all contributions (black), (ii) omitting the change in ozone (red), (iii) omitting the change in terpene emission (green) and (iv) omitting the suppressing effect of water vapour (blue).

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From that the statement of the role of terpenes becomes apparent. Omitting the emission change would result in an unchanged parameter as can be considered for NP A. Ozone is certainly important to and a possible future situation with less water (water stress conditions) would cause the highest effect. Since the NP represent the formation of new particles and correlate with the concentration of new aerosols, a rise in any NP would result in a higher nucleation probability and aerosol number concentration in the future.

If we consider the expected climate warming with an average of +3 K (IPCC report, 2007), which is at the lower limit for boreal regions, we obtain a rise in nucleation event number between +12% ($T_{ref}=0^{\circ}\text{NP}$) and +30% ($T_{ref}=20^{\circ}\text{NP}$), which is substantially. A temperature increase by +6 K would result in an increase between 32 and 71%. If sesquiterpenes participate in nucleation the increase is expected to be even higher because of the stronger temperature dependency of the emissions. On the other hand the number of events would drop only if ozone is reduced remarkably to compensate the rise in terpene and water vapor concentration. According to the conclusions of the IPCC report (2007) this is very unlikely.

5 Conclusions

We have postulated a new nucleation parameter to describe the occurrence of ambient particle formation at boreal and coniferous forest areas, which uses ozone and water vapor as proxies for the contribution of reactive organic trace gases and UV B radiation strength for OH production. The parameterisation is found capable in explaining seasonal and annual variation of observed nucleation events at different sites. Using this parameter it seems very likely that nucleation events will increase in number over the entire boreal regions and several mid-latitude areas having consequences on aerosol and cloud properties. It seems a step further in understanding biosphere-atmosphere cloud climate feedback processes (Kulmala et al., 2004b) initiated by the biosphere to protect the biosphere from harm. A rise in aerosol number and thus cloud condensa-

tion nuclei will initiate cloud formation and might increase the lifetime of clouds which subsequently cools the Earth's surface. However to exactly predict the future rise and intensity of nucleation events a notably improved knowledge about terpene emissions, future ozone, carbonyl compounds and sulfuric acid is essential.

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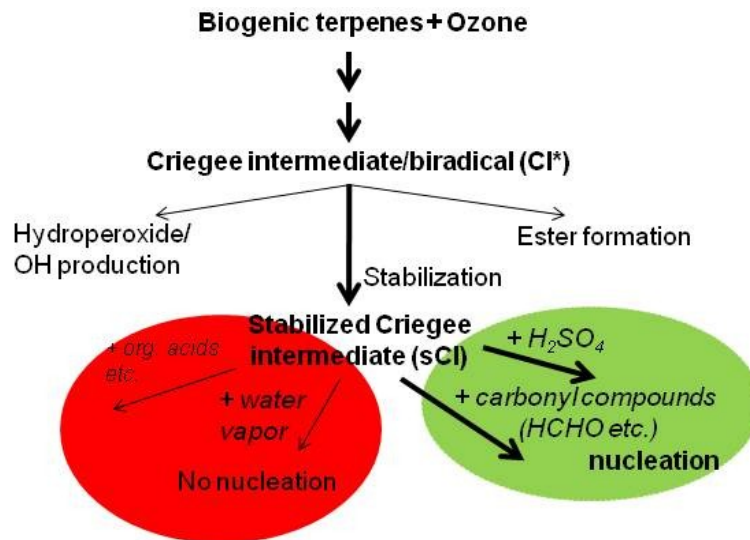


Fig. 1. Schematic representation of the nuclei formation. Highlighted in red are suppressing reactions, and in blue reactions leading to nucleation.

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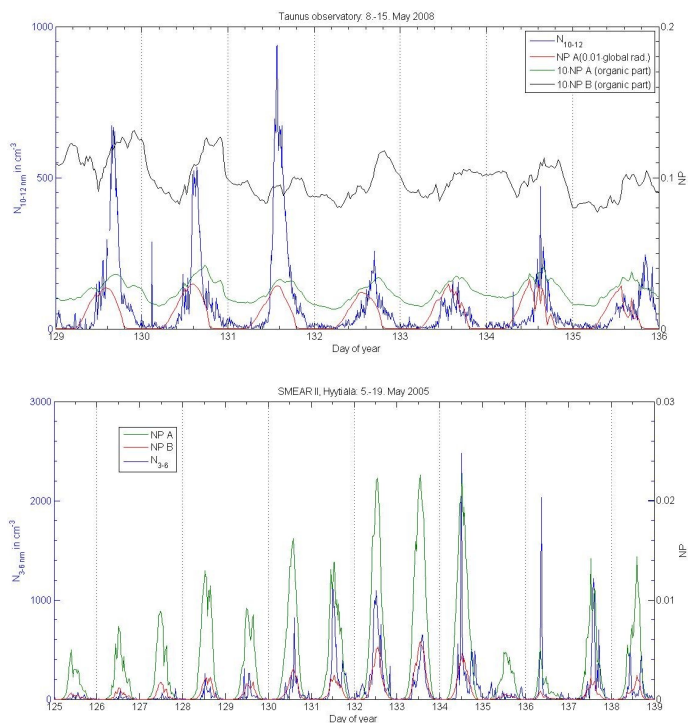


Fig. 2. Intercomparison of nucleation parameters and total number concentration of aerosols between 10 and 12 nm in diameter at the German site (top) or between 3 and 6 nm in Finland (bottom). Since no UV B measurements are available at the German site, UV B was exemplarily replaced by 1% of the global radiation.

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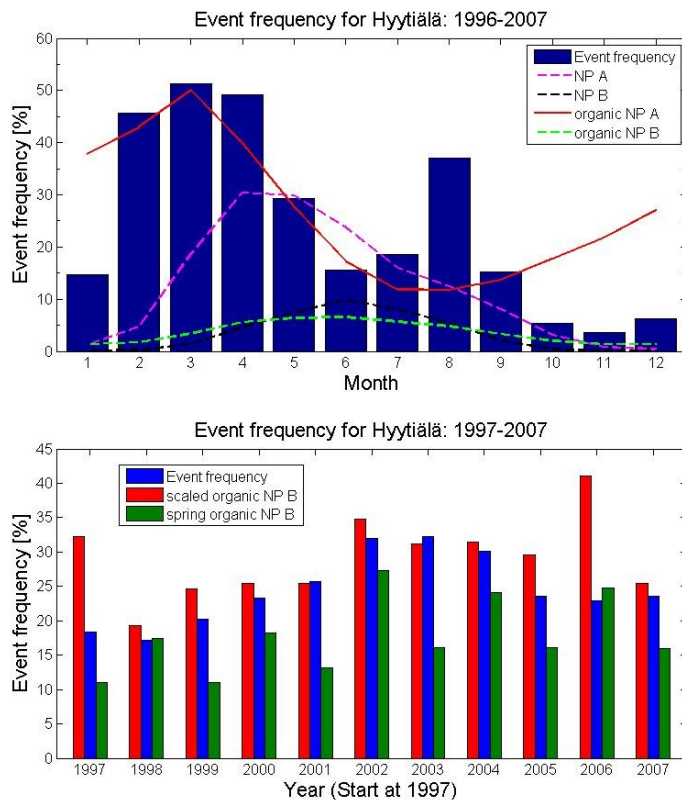


Fig. 3. Intercomparison of nucleation parameters and total number concentration of aerosols between 10 and 12 nm in diameter at the German site (top) or between 3 and 6 nm in Finland (bottom). Since no UV B measurements are available at the German site, UV B was exemplarily replaced by 1% of the global radiation.

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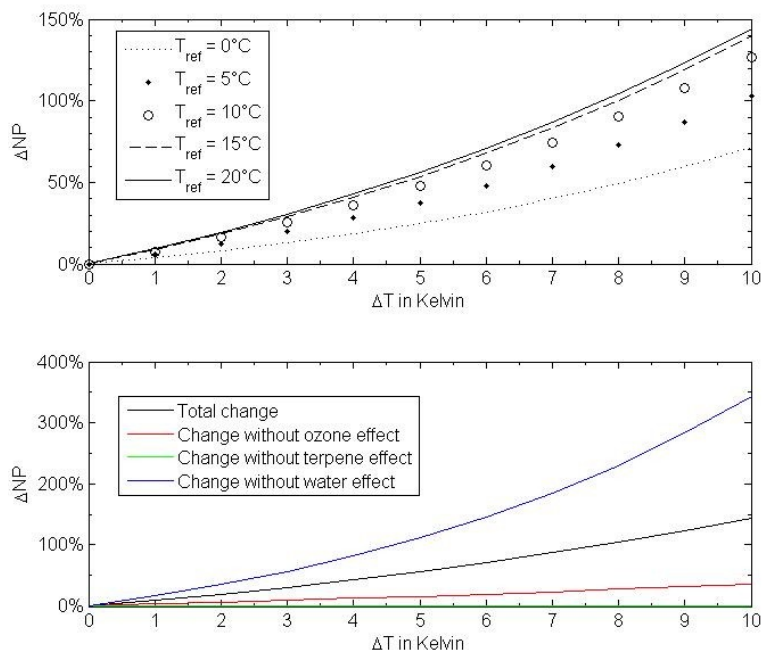


Fig. 4. Displayed is the calculated relative change in NP because of changes in ozone, temperature and water vapour. Top: Relative changes of NP at different reference temperatures due to a change between 0 and 10 K. Bottom: Relative changes of NP at a reference temperature of 20°C of the individual changes of different parameters.

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