# Parameterization of particle formation rates in distinct atmospheric environments

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**Abstract.** Atmospheric particle formation rate (*J*) is one of the key characteristics in new particle formation (NPF) processes worldwide. It is related to the development of ultrafine particle growth to cloud condensation nuclei (CCN) and, hence, to Earth radiative forcing in global models, which helps us to better understand the impact of NPF on cloud properties and climate change. In this work, we parameterized four semi-empirical *J* models for 5 nm atmospheric particles using field measurements obtained from distinct environments that varied from clean to heavily polluted regions and from tropical to polar regions. The models rely primarily on sulfuric acid as a condensing vapor, condensation sink to account for the vapor loss, and relative humidity for meteorological contribution to *J*. However, the dependencies between *J*, condensation sink, and relative humidity are affected by their interlinked relations to sources and sinks of other condensable vapors than sulfuric acid and the potential traffic emissions to the observed size range. The parameterization results showed that our models were able to produce plausible predictions for boreal forest environments, heavily polluted environments, and biogenic environments with high relative humidity. We further tested the models in the global simulation module Tracer Model 5 (TM5, massive parallel version) to simulate particle number size distribution across 14 global atmospheric

measurement sites. The simulated results showed satisfactory predictions on particle number concentrations for all the tested environments, with significant improvement in the nucleation mode, and better prediction accuracy for Aitken and accumulation modes compared to the binary sulfuric acid-organic vapor model in Riccobono (2014). Our study has successfully provided powerful tools to predict $J_5$ on a global scale across various environment types using the most essential and more accessible variables involved in the NPF processes. Essentially, this work reinforces the necessity for global research into the investigation of environment-oriented meteorology-involved NPF processes.

## 1 Introduction

Atmospheric new particle formation (NPF) is a natural phenomenon observed globally (Bousiotis et al., 2021; Brean et al., 2023; Gordon et al., 2017; Kerminen et al., 2018; Nieminen et al., 2018). As particles form and grow on regional scales, they can reach large enough sizes at which they can act as cloud condensation nuclei (CCN) for water vapor to condense onto when forming clouds. This process affects cloud properties (Roldin et al., 2011; Sanchez et al., 2016; Spracklen et al., 2008) and ultimately the global climate depending on the particle numbers, sizes and their chemical compositions (Bellouin et al., 2020; Calvo et al., 2013; Uno et al., 2020). Particle formation rate ($J$) is an essential parameter describing the NPF intensity, which is often utilized to represent NPF in global models to simulate the effect of NPF on cloud properties and radiative forcing on Earth. To derive a representative parametrization of $J$ for global simulation, we require a broad understanding of NPF in different environments temporally and spatially. That being the case, the atmospheric measurements of both particle number size distributions and NPF precursor vapors are essential to obtain for the atmospheric observations and model developments.

The NPF processes have been investigated and parameterized based on particle formation mechanism theories (Chang et al., 2009; Kulmala et al., 2001; Lehtinen and Kulmala, 2003), field measurements from specific environments, such as pristine boreal forests (Kulmala et al., 2001; Nieminen et al., 2011; Paasonen et al., 2010), urban cities (Salma et al., 2011, 2016, 2021; Salma and Németh, 2019; Zhang et al., 2010), rural area (Lee et al., 2019; Yli-Juuti et al., 2009), marine environment (Zhang et al., 2010), and also chamber experiments (Kirkby et al., 2011; Lehtipalo et al., 2018). In addition to neutral particle formation mechanisms, the ion-induced nucleation is also covered in $J$ parameterizations (e.g. Nieminen et al., 2011; Määttänen et al., 2018). The existing literature primarily focused on the particle activation and survival of nucleation mode particles down to 1.5 nm, involving complex micro-physics of aerosol particles, such as nanometer clusters production, losses due to clusters coagulation and growth (Bousiotis et al., 2021; Chu et al., 2019; Kerminen et al., 2018; Nieminen et al., 2018). Furthermore, distinct effects on particle formation rates influenced by the same factors were seen under comparable environmental settings. However, these environment-specific models typically have limited application for global simulation implementations that encompass the diverse atmospheric conditions on Earth.

To simulate particle formation rates, one usually starts from the nucleation mode size range. Global modelers have been facing great challenges in simulating nucleation mode particles because large-scale models have limited capabilities in treating the complicated aerosol dynamics taking place in the sub-5 nm particle size range. The formation rate at 5 nm is shown to be important because after sizes of about a few nm in diameter, particle growth rates show relatively limited variability in different environments (Kulmala et al., 2022a, b, 2023). In addition, we currently do not have a good enough theoretical understanding on the processes dictating particle growth rates at the smallest sizes, nor the survival of such particles from coagulation scavenging (e.g. Cai et al., 2022; Tuovinen et al., 2022; Marten et al., 2022).

The essential parameters for $J$ parameterizations should include at least one type of precursor vapor, some may also cover meteorological parameters, and the sinks for vapors and particles. For instance, sulfuric acid ($H_2SO_4$) as the most known precursor vapor plays a critical role in particle formation and growth processes due to its low volatility (Kulmala et al., 2004; Myllys et al., 2019). In the earlier parameterizations of NPF mechanism, $J$ correlated (linearly or squared) with $H_2SO_4$ concentrations in various environments (Paasonen et al., 2010). In terms of meteorology, air temperature ($T$), relative humidity (RH), global solar radiation (GRad), wind speed (WS), and wind direction influence the particle formation rates in certain environments as well (Laarne et al., 2022; Salma et al., 2021; Zaidan et al., 2018). The variation of $T$ can influence the precursor vapor formation and stability of NPF processes: a higher $T$ can enhance the biogenic emissions that participate in particle formation in a boreal forest (Dada et al., 2017; Nieminen et al., 2015), while a lower $T$ favored the $H_2SO_4$-amine clusters stability in a megacity (Deng et al., 2020). RH can impact the precursor vapor formations as well as the aerosol formation rates (Ding et al., 2021; Hellmuth, 2006). The variation of RH is dependent upon $T$ so that the rise of $T$ during daytime increases the planetary boundary layer height (PBLH), which in turn dilutes the air mixture and decreases the RH (Liu et al., 2018) as well as particle number concentrations (Mazon et al., 2016) in the atmosphere. For condensable vapor loss, we usually include the term condensation sink (CS), which describes the loss rate of condensable vapors to aerosol particles, and it typically declines before an NPF event starts. $H_2SO_4$ concentrations, on the other hand, increase due to the reduction in CS, which means that the condensable vapors are not lost onto the aerosol particles as efficiently as they would be at greater CS values (Hellmuth, 2006; Kulmala et al., 2012).

In general, the developed $J$ models underestimate the observed particle number concentration, which may be attributed to NPF schemes being poorly represented in these models. Many $J$ parameterization works were conducted focusing on the formation mechanisms from sulfuric acid (Paasonen et al., 2010), sulfuric acid-water (Määttänen et al., 2018), sulfuric acid-ammonia (Glasoe et al., 2015), and sulfuric acid-organic vapor (Paasonen et al., 2010; Riccobono et al., 2014). However, some models are likely applicable only to certain types of environment, or they primarily cover the microphysics of the particle nucleation at sub-3 nm range, where the nucleated clusters face higher instability due to the higher evaporation rates than condensation rates (Deng et al., 2021; Wang et al., 2011). Bergman (2022) attempted an organic-vapor-based NPF scheme in addition to the commonly used binary water-sulfuric-acid-based scheme to simulate global particle formation and number concentrations. This scheme improved the simulated number concentrations across the observation stations, although they were still underestimated compared to the observations, suggesting that the parameterization of early growth of particles to 5 nm diameter still requires improvement.

To predict the particle formation rate at 5 nm originating from NPF and subsequent growth, as well as to understand and predict the climatic impacts caused by NPF and initial growth in global scale, we parameterize particle formation rates ($J$) at 5 nm using combined measurement data from six different environments: Hyytiälä (boreal forest close to rural environment, Finland), Beijing (megacity, China), Värriö (remote boreal forest, Finland), Budapest (urban, Hungary), Agia Marina Xyliatos (rural, Cyprus) and Manacapuru (Amazonian basin, Brazil). The parameterizations of $J$ were based on the analysis of atmospheric particle number-size distributions. Sulfuric acid concentrations, RH and CS are the main input variables in the parameterization models. By including information from various types of environments, we will be able to demonstrate whether our models can adequately explain the formation rate of 5 nm particles on a wider environmental scale. The parameterized models are then incorporated into EC-Earth models to simulate particle formation rates in the global scale (the European community Earth-System Model, EC-Earth, chemistry transport model TM5: Tracer Model 5, version TM5-chem-v3.0, details in supplement) (Huijnen et al., 2010).

This work aims to provide an effective tool for global particle formation rate estimations. Our parameterizations have three main features: (1) the number of inputs is limited to be the most essential parameters involved in NPF process, (2) they do not involve complex microphysics at particles smaller than 5 nm, and (3) they cover a wide range of environment types. These features will enhance the applicability of the parameterizations for the purpose of global model application.

## 2 Measurement locations and instrumentation

This study includes measurements from six different sites representing different environmental conditions. A summary for all locations and the instrumentation used is given in Table S1. Figure 1 shows the map of the measurement sites included in this study.

### 2.1 Measurement sites

#### 2.1.1 Rural boreal forest environment: Hyytiälä, Finland

The measurement data were obtained from the SMEAR II-station (Station for Measuring Ecosystem–Atmosphere Relations), situated in a Scots pine (*Pinus sylvestris*) forest in Hyytiälä (61.1 N, 24.17 E; 181 m (a.s.l).; Hari and Kulmala, 2005), southern Finland. This measurement site is described as having a rural regional background with minimal anthropogenic emission. Hyytiälä data covered the period from 21 March 2016 to 18 August 2019.

#### 2.1.2 Remote sub-arctic boreal forest: Värriö, Finland

The SMEAR I measurement station (67°45′31° N, 29°36′41° E, 390 m a.s.l.) was built on the top of Kotovaara hill located in north-eastern Finland. Similar to Hyytiälä, the site is also a rural background covered mainly by Scots pine (*Pinus sylvestris*) forest located at the north side of the Värriö fell range. However, it is affected by potential polluted airmass that comes from Kola peninsula rather than local industrial pollutants. A detailed description of SMEAR I station can be found in (Kyrö et al., 2014). The data used from Värriö were from 5 April to 13 August 2019.

#### 2.1.3 Polluted megacity: Beijing, China

In Beijing, the measurements were performed at the west campus of the Beijing University of Chemical Technology (BUCT, 39.94° N, 116.30° E, 20 m a.s.l.). The sampling took place from outside the window on the 5$^{th}$ floor of the university building close to a street with busy traffic. For more details on the description of BUCT measurement site, see Liu et al., 2020. The data were available from 29 May 2018 to 3 April 2019.

#### 2.1.4 Urban site: Budapest, Hungary

The measurements took place at the Budapest platform for Aerosol Research and Training (BpART) Laboratory (47.47° N, 19.06° E; 115 m a.s.l.) of the Eötvös Loránd University situated on the bank of the river Danube. The site represents a mixed average atmosphere of the city center (Salma et al., 2016). The data were obtained from 22 March to 17 April 2018.

#### 2.1.5 Mediterranean rural site: Agia Marina, Cyprus

The measurements were conducted at the Agia Marina Xyliatou (AMX) station (35.03° N, 33.05° E; 532 m a.s.l.) of the Cyprus Atmospheric Observatory (CAO). The site represents a rural background location situated at the foothills of Troodos mountains, with agriculture land in the vicinity. The data were obtained between 22 February and 3 March 2018. For more details about the site, see e.g. Baalbaki et al., (2021).

#### 2.1.6 Amazonian basin: Manacapuru, Brazil

The Manacupuru measurement site was in a pastureland 70 km west of Manaus, Brazil, in central Amazonia. This site receives airmass from various resources, including rural, biogenic and anthropogenic from the nearby municipality (Manaus). The trace gases and meteorological measurements were performed during the GoAmazon2014/5 campaign at the T3 site (3.2133◦ S, 60.5987◦ W 50 m a.s.l.), 10 km northeast of Manacapuru, Brazil (Martin et al., 2016; Schiro et al., 2018). A more detailed description of the measurement site can be found in (Myers et al., 2022). The data covered the time period from 22 August 2014 to 9 October 2014.

### 2.2 Instrumentation

#### 2.2.1 Sulfuric acid measurements and proxies

$H_2SO_4$ concentrations were measured at all sites, except for the Amazonian basin, using a Chemical Ionization Atmospheric Pressure interface Time-of-Flight spectrometer (CI-APi-ToF) (Eisele and Tanner, 1993; Jokinen et al., 2012) with $NO_3^-$ as the reagent ion and analyzed using tofTools package based on MATLAB software (Junninen et al., 2010). In the Amazonian basin, $H_2SO_4$ concentrations were measured using a selected ion chemical ionization mass spectrometer (SICIMS), see Myers et al., (2022) for more details. The $H_2SO_4$ concentration measurements were taken from different levels ranging from ground level up to 35 meters above ground level. The CI-APi-ToFs were calibrated uniformly before the measurement in each location following the technique described by (Kürten et al., 2012), except for the Amazonian basin where the selected ion chemical ionization mass spectrometer (SICIMS) was calibrated following the scheme described in Mauldin III et al. (1998).

To increase the applicability of our derived parameterization, $H_2SO_4$ proxy data from Hyytiälä and Beijing were included as an additional testing data set. The proxy data were calculated using the proxy specific for the boreal forest environment and polluted megacity developed by Dada et al., (2020). For Hyytiälä, the sulfuric acid proxy data ranged from 22 August to 25 December 2016, and 8 March 2018 to 26 February 2019, denoted as $\text{Hyytiälä}_{\text{SAprx}}$; For Beijing, the time period was from 15 March to 3 April 2019, denoted as $\text{Beijing}_{\text{SAprx}}$. The subscript "SAprx" (SA as in sulfuric acid) in $\text{Hyytiälä}_{\text{SAprx}}$ and $\text{Beijing}_{\text{SAprx}}$ indicates that the datasets utilize $H_2SO_4$ concentration from proxies as input for the testing dataset.

### 2.2.2 Particle number size distribution

The particle number size distribution (PNSD) measurements were obtained from different types of setups in each site. Hyytiälä: twin-Differential Mobility Particle Sizers (DMPS; Aalto et al., 2001); Värriö: Differential Mobility Particle Sizers (DMPS) (Jokinen et al., 2022); Beijing: Particle Size Distribution (PSD) system with a nano-Differencial Mobility Analyzer (DMA) and an Aerodynamic Particle Sizer (APS) (Zhou et al., 2021); Budapest: flow-switching-type DMPS (6 – 1000 nm; Salma et al., 2016); Cyprus: Neutral cluster and Air Ion Spectrometer (NAIS) and Scanning Mobility Particle Sizer (SMPS; Baalbaki et al., 2021); Amazonian basin: the measurements were conducted using SMPS (10 – 1000 nm). It is important to note that we do not aim to compare the PNSD measurements from all the chosen sites. Instead, the PNSD measurements were used to calculate the formation rates based on changes in particle number concentrations under local conditions.

### 2.2.3 Meteorological variable

The meteorological variables included in this study are relative humidity (RH, %) and ambient temperature ($T$, ºC). In Hyytiälä, RH and $T$ were measured at 16.8 m using Rotronic MP102H RH sensor (Rotronic Hygromet MP102H with Hygroclip HC2–S3, Rotronic AG, Bassersdorf, Switzerland); In Värriö, RH and $T$ were measured by a Rotronic MP106A captive sensor; In Beijing, RH and $T$ were monitored by Vaisala weather station (AWS310); In Budapest, RH and $T$ were monitored using Vaisala HMP45D temperature and humidity probe, and Vaisala WAV15A anemometer located on on-site of the BpART Lab; In Cyprus, RH and $T$ were measured by a meteorological station in a nearby village (35.01◦ N, 33.05◦ E), 2.85 km away from the measurement site; In Amazonian basin, RH and $T$ were measured at the Atmospheric Radiation Measurement (ARM) user facility.

## 2.3 Data analysis

### 2.3.1 Calculation of particle formation rates

To develop more inclusive and generalized models, the parameterization included data from both NPF event days and non-event days. This approach recognizes that the production of atmospheric secondary particles from non-NPF events (days with no apparent particle growth) is becoming more significant in a world with growing anthropogenic influence (Kulmala et al., 2022a). Such a measure would increase the applicability of our models on a global scale.

The observed particle formation rates ($J_5$) at 5 nm were calculated from the measured PNSD according to Equation 1 (Kulmala et al., 2012).

$$J_{d_p} = \frac{dN_{d_p}}{dt} + CoagS_{d_p} * N_{d_p} + \frac{GR}{\Delta d_p} * N_{d_p} \qquad \text{(Eq. 1)}$$

The first term $dN_{dp}/dt$ is the change in concentration in the size bin, 5–9 nm. Ideally, this term, as well as the concentration of particles within the size range, $N_{dp}$, in the following terms, are associated with the growth of particles formed by atmospheric NPF past 5 nm; however, especially in traffic-related environments, they may also have an unknown contribution by direct particle emissions to this respective size range (Okuljar et al., 2021; Rönkkö et al., 2017). The second term $CoagS_{dp}$ is the coagulation sink, which describes the 5-9 nm particle losses due to coagulation with larger particles calculated from the PNSD at each measurement site (Kulmala et al., 2012). The third term describes the loss of particles due to their growth out of the size bin. Here, we calculated the growth rates (GR) of 5–9 nm particles using the maximum concentration method (Kulmala et al., 2012) for days classified as NPF event days as described by Dal Maso et al., (2005). The GR for non-event days was approximated using the normalized PNSD from the sum of non-NPF events at each site. Such approximation is validated for several locations as a ‘quiet NPF’ occurs with the similar GR as that on NPF event days (Kulmala et al., 2022a).

### 2.3.2 Extrapolation of particle formation rates

For Budapest and the Manacapuru (Amazonian basin), the particle formation rates were calculated from PNSD measurements at 6 nm and 10 nm, respectively. Therefore, we obtained $J_5$ by extrapolating from $J_6$ and $J_{10}$ respectively. The $J_5$ extrapolation followed the analytical formula derived by Kerminen and Kulmala (2002). We extrapolated $J_5$ from $J_6$ for Budapest. For Manacapuru, the extrapolations were done separately for $J_{10}$ (wet season) and $J_{14}$ (dry season), due to the particle size limit of the measurement instrument.

### 2.3.3 Condensation and coagulation sink (CS and CoagS)

The CS and CoagS were calculated from the measured PNSD data for each site using the method proposed by Kulmala et al., (2012). To ensure the comparability between all locations, both CS and CoagS were calculated without the correction for hygroscopic growth. There are several ways to determine the hygroscopic growth factors in CS and CoagS calculations. Laakso et al., (2004) developed parameterizations for Hyytiälä solely based on the meteorological conditions and the aerosol composition in Hyytiälä, which results in the inapplicability of that method to other sites. In the Supplementary of Baalbaki et al., (2021), Figure S4 shows the CS with hygroscopic correction is about 1.1 – 1.3 times higher than dry CS, which would result in an overestimation on CS for the case in Cyprus. Petters and Kreidenweis (2007) introduced the single hygroscopicity parameter κ (kappa), which can be derived from Humidified Tandem Differential Mobility Analyzer (HTDMA) or cloud condensation nuclei counter measurements or based on aerosol chemical composition obtained from instruments such as the Aerosol Chemical Speciation Monitor (ACSM) or Aerosol Mass Spectrometers (AMS). In other locations, since organics are typically the dominant component of aerosol mass in continental areas or marine polluted areas (Chen et al., 2022) and are less hygroscopic than inorganics, one can expect an underestimation of CS similar to the one reported in (Baalbaki et al., 2021). As a result, we omitted the hygroscopic growth impact for the chosen measurement sites to harmonize the data composition and the later model analysis.

### 2.3.4 Datasets

The parameterizations were developed using the combined dataset from all six measurement sites in hourly time resolution. Data points were selected considering detection limit of the instruments and therefore, the filters were set to be $J_5 > 1\times10^{-5}$ $cm^{-3}$ $s^{-1}$, $H_2SO_4$ concentration $> 5\times10^3$ $cm^{-3}$, RH $\in [0,100]$ % and CS $> 1\times10^{-5}$ $s^{-1}$. The complete dataset was afterwards randomly resampled into a training set (75% from the complete dataset) and a testing set (25% the rest of the complete dataset) for parameterization. In model testing, we included two additional inputs from $H_2SO_4$ concentration proxies developed by Dada et al., (2020) from Hyytiälä and Beijing. The detailed number of data points per site are shown in Table 1. The data distribution and comparison of each input variable are displayed in Figure S2, where the overall variations of the input variables across the six sites are distinct in their range and intensity, which pronounces the inclusivity of model training for a wider application in global environments.

## 3 Parameterization of $J_5$

### 3.1 Derivation of parameterization models

We derived the parametrized $J_5$ based on the input variables ($H_2SO_4$, RH, CS), which were chosen based on field observations that highlighted their roles in the particle formation mechanism across various environments (Baalbaki et al., 2021; Dada et al., 2020; Kerminen et al., 2018; Myers et al., 2022; Salma et al., 2016, 2021; Yan et al., 2021). It has been discovered that NPF events occur favorably under lower RH, for example in boreal forests (Dada et al., 2018; Yao et al., 2018), Mediterranean regions (Debevec et al., 2018), from CLOUD chamber experiment (Duplissy et al., 2016) and model studies (Hamed et al., 2011). RH was shown to be seasonally related to cloudiness and global radiation, so that a decreasing global radiation can lead to an increased RH and cloudiness within the troposphere (Ruosteenoja and Räisänen, 2013). To reduce the model complexity, we opted to use RH as an indirect indicator of global radiation. A lower CS facilitates the occurrence of NPF events even in contrasting environments with distinct types of condensable vapor. For example, CS is a measure of a sink for anthropogenic vapors in a megacity (Wang et al., 2011) and for biogenic vapors in a clean boreal forest (Dada et al., 2017; Tuovinen et al., 2020), as well as a sink for growing sub-5 nm clusters and particles (Kulmala et al., 2017). When combined with $H_2SO_4$ as an input variable, the evidently important sink effect of a pre-existing particle population on the ambient $H_2SO_4$ concentration is implicitly transferred from CS to $H_2SO_4$ in our parameterization. Indirectly, CS may also be associated, either causally or not, with 1) emissions or sinks of vapors other than $H_2SO_4$ participation in NPF or particle growth, and 2) primary particle emissions from traffic, which would influence particle formation rates estimated from observations using eq. 1. Furthermore, since we omit the influence of hygroscopic growth of particles on CS, a fraction of real sink effect of CS is implicitly transformed to the variable RH in our parameterization.

We tested with $T$ as an input variable during model derivation and training. However, the modelled results did not show improvement compared to the current parameterization, suggesting that $T$ provided redundant information for describing particle formation in the context of our model's global application.

Other than sulfuric acid, highly oxygenated organic molecules (HOMs) and ammonia ($NH_3$) have been discovered to play a significant role in particle formation process (Bianchi et al., 2019; Lehtipalo et al., 2018). The possible cluster types may include $H_2SO_4$-$NH_3$-$H_2O$ (Yu et al., 2018) and $HNO_3$-related clusters such as $HNO_3$-$H_2SO_4$-$NH_3$ in the upper tropospheric particle nucleation (Wang et al., 2020, 2022, 2023). Yet, we are unable to include HOM nor $NH_3$ concentrations owing to limited data availability from the chosen measurement sites. So far, long-term measurements (> 1 year) of HOMs, matching the time range covered by other variables, are only available in Hyytiälä from a CI-APi-ToF mass spectrometer. However, this is not the case at other sites, limiting our ability to have simultaneous HOMs data across all environments included in this study. Similarly, the $NH_3$ concentrations either did not cover the same time period as other variables or were unavailable for the other environments.

#### 3.1.1 Different versions of the parameterization models

The derived model functional forms are as follows:

Model 1 (the baseline model, Eq. 2) presents the simplest particle formation mechanism based solely on the abundance of the precursor vapor $H_2SO_4$ concentrations in the atmosphere. The coefficient $k_1$ serves as a scaling coefficient that represents the activation rate of clusters in the presence of $H_2SO_4$ molecules during cluster formation (Kulmala et al., 2006; Paasonen et al., 2010).

$$J_5 = k_1 \times [H_2SO_4] \qquad \text{(Eq. 2)}$$

Model 2 (Eq. 3) introduces RH in addition to model 1 to partially represent the effect of the changing meteorological conditions relating to the global radiation and ambient water vapor content on $J_5$ in general in different types of environments (Dada et al., 2017; Hamed et al., 2011; Li et al., 2019). The coefficient $k_2$ serves as a scaling coefficient and shown as the activation efficiency of the nucleated clusters.

$$J_5 = k_2 \times [H_2SO_4] \times RH^{k_{RH}} \qquad \text{(Eq. 3)}$$

Model 3 (Eq. 4) includes, in addition to model 2, the factor CS. As discussed above, in our parameterization CS is connected not only to the sink of newly formed particles prior to their growth past 5 nm, but possibly also to sinks or sources of vapors other than $H_2SO_4$ participating in particle formation and growth and, in polluted environments, to sub-10 nm particle emissions from traffic. The coefficient $k_3$ serves as a scaling coefficient for the activation and survival efficiency of the nucleated clusters.

$$J_5 = k_3 \times [H_2SO_4] \times RH^{k_{RH}} \times CS^{k_{CS}} \qquad \text{(Eq. 4)}$$

Model 4 (Eq. 5), additionally accounts for the formation of $H_2SO_4$ multimers in the gas phase prior to cluster formation as assumed by the kinetic theory (McMurry and Friedlander, 1979), the coefficient $k_{SA}$ represents the number of $H_2SO_4$ molecules (2, 3, 4, etc…). Therefore, $k_4$ in this case is not the activation coefficient anymore but includes both the collision frequency and the probability of a stable particle formation after the collision (Sihto et al., 2006; Weber et al., 1996).

$$J_5 = k_4 \times [H_2SO_4]^{k_{SA}} \times RH^{k_{RH}} \times CS^{k_{CS}} \qquad \text{(Eq. 5)}$$

### 3.2 Model training results

To derive a parametrized $J_5$ based on precursor and other input variables from the training dataset, we used the "*fmincon*" optimization algorithm in MATLAB to retrieve the values of each coefficient ($k_1$-$k_4$, $k_{SA}$, $k_{RH}$ and $k_{CS}$) from the training datset. The coefficients obtained for each of the models can be found in Table 2. The derived models with the optimized coefficients were applied to the testing datasets and compared with the observed $J_5$ and the parametrized $J_5$. We evaluated the performance of each model based on the data distribution, the resulting deviation from observation and its uncertainty. To maintain the global model's simplicity, the parameterization covered both daytime and night-time data for all sites in all models.

Figure S3 presents the measured to modelled $J_5$ from model 1-4 using training dataset from six measurement sites, including the slopes and coefficient of determination ($R^2$). Overall, by comparing model 1 (Fig. S3a) and model 2 (Fig. S3b), we observed an improvement in the model performance with the inclusion of RH. The $R^2$ value improved from 0.28 to 0.44, and the slope increased from 0.29 to 0.56. This observation confirmed the importance of considering meteorological impact when parameterizing $J_5$. By further including CS in model 3, the model improved further (Fig. S3c), with the $R^2$ increasing from 0.44 to 0.49, and the slope from 0.56 to 0.62. To further introduce the kinetic theory and the formation of $H_2SO_4$ dimers and other multimers, we added an exponent over $H_2SO_4$ in model 4 (Fig. S3d). This addition showed a further improved correlation and slope between the measured or modeled data for the training datasets ($R^2$ = 0.57, slope = 0.76). In subsequent testing, model 4 generally outperformed the other models (see section 4, Fig. 2).

### 3.3 Model evaluations

#### 3.3.1 MAE and RMSE

We computed the mean absolute errors (MAE), root mean square errors (RMSE) for each model using the testing dataset to gain a better understanding of the models' performance. The numerical values of MAE and RMSE are given in Table S3.

The MAE calculation equation is as follows:

$$MAE = \frac{1}{n}\sum_{i=1}^{n}|y_i - \hat{y_i}| \qquad \text{(Eq. 6)}$$

where $n$ is the number of data points (here it is the total number of data points from testing set, see Table 1), $y_i$ is the observed value, and $\hat{y_i}$ denotes the predicted value. MAE measures the accuracy of models' prediction power, by quantifying the average magnitude of errors between observed and predicted values (Chai and Draxler, 2014). A lower model error is manifested by a lower MAE value.

The RMSE is calculated as the square root of the difference between the measured $(y_i)$ and predicted $(\hat{y_i})$ $J_5$ values normalized by the number of data points.

$$RMSE = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(y_i - \hat{y_i})^2} \quad \text{(Eq. 7)}$$

RMSE also measures the average magnitude of errors of models. However unlike MAE, RMSE squares the errors giving greater weight to larger errors and penalizing them more heavily (Chai and Draxler, 2014). Therefore, RMSE values reveal whether the models' performances are highly influenced by large prediction errors Similar to MAE, lower RMSE values indicate better model performance.

Figure S4 (upper panel) depicts a declining trend of the overall MAE from models 1 to 4 (Eqs. 2-5). For the environmental types investigated in this study, the MAE values of the four models from all sites are lower than 1, indicating that the mean differences on the magnitude for $J_5$ are minor when utilizing the parameter settings from our models. However, Budapest stands out due to the apparent higher MAE, potentially highlighting the distinct NPF mechanism in Budapest compared to the other sites as well as the seasonal limitations on its data (spring 2018 only).

The RMSE values increased as more parameters were added into the model, peaking for model 3 (Fig. S4, lower panel), even though model 3 can predict $J_5$ for multiple types of environments on a satisfactory level. We can see that from model 1 to model 2, the inclusion of RH increased the model errors more compared to the addition of CS from model 2 to model 3. However, the RMSE values dropped significantly when $H_2SO_4$ was allowed to vary with an exponent $k_{SA}$ in model 4 in the presence of both RH and CS.

Based on the results summarized above, models 3 and 4 (Eqs. 4 & 5) seem to be the most promising for global $J_5$ prediction among all model types owing to their low MAE values. However, the lower RMSE for model 4 showed its outperformance to model 3.

### 3.3.2 Akaike Information Criterion

The Akaike Information Criterion (AIC) is a statistical measure that helps to evaluate the goodness-of-fit of a statistical model. We use AIC as an evaluation tool because it can evaluate models with different number of parameters and complexities, ensuring a balanced assessment. Eventually, it allows us to select the model with the best balance between the model complexity and goodness-of-fit. The parameters used to calculate the AIC for each site are shown in Tables S4-S6. A lower AIC score indicates a superior goodness-of-fit and a lower tendency for model overfitting. The relative likelihood term ($L$, $L = e^{(\mathrm{AICmin}-\mathrm{AICi})/2}$), calculated from AIC scores, reflects the likelihood that the *i*th model minimizes information loss as compared to the model with the lowest AIC. A relative likelihood of 1 suggests that the model outstands other models in minimizing information loss. For boreal forest environments (Table. S4), and urban environments (Table. S5), both models 1 and 4 minimized information loss the most. For rural regions, model 4 (Eq. 5) performs the best (Table. S6). Compared to the baseline model (model 1, Eq. 2), we find that $H_2SO_4$ is a more powerful parameter than RH or CS in all environments. However, in Manacapuru, including RH and CS shows clearly an improved predictive accuracy in model 4 (Eq. 5).

## 4 Results and discussion

### 4.1 Parameterization testing results

The scatterplots (Fig. 2) demonstrate the overall performance of the parameterizations from the 4 models (Eqs. 2-5) using the testing dataset. The overall and site-specific Pearson's coefficients, slopes from robust linear fit between the measured and modelled $J_5$, as well as the number of data points from the testing dataset, can be found in Table 3. Overall, $r$ increased significantly for model 2 and 3 (Fig. 2b & 2c, $r$ = 0.69, $r$ = 0.71) compared to model 1 (Fig. 2a, $r$ = 0.55) as we include relative humidity and condensation sink as model parameters. Model 4 provides the best linear fit results, implying that the model can predict an overall reliable estimation on $J_5$ in all the investigated environment types (Fig. 2d, $r$ = 0.78). It is notable that with the combined data sets, the condensation sink receives a positive exponent in models 3 and 4 ($k_{CS}$ = 0.56 and 0.67, respectively), likely due to its association with concentrations of other condensable vapors than $H_2SO_4$ and traffic emissions.

### 4.1.1 Boreal forests: Hyytiälä and Värriö

Given the boreal forest background, Hyytiälä and Värriö exhibited comparable variations in the distribution of modelled $J_5$ values from the four model types. As shown in Figure 3, model 3 ((a3), (b3), (c3)) and model 4 ((a4), (b4), (c4)) illustrated a more centered data distribution between the modelled and measured $J_5$, which demonstrates a potential favoring of NPF under low RH conditions, likely associated with increased global radiation for the boreal forest environment (Dada et al., 2018; Hamed et al., 2011) and lower sinks for vapors and growing sub-5 nm particles (see Section 3.1). Notice that the mean $H_2SO_4$ concentration in Värriö is about twice as high as that in Hyytiälä, opposite to CS which is clearly lower in Värriö (Table S2). The low CS in Värriö compared with Hyytiälä is primarily due to the lower emission rate of the regional precursor vapors (e.g. Tunved et al., 2006), leading also to the lower observed NPF event frequencies (Kyrö et al., 2014; Neefjes et al., 2022). We must note that the Hyytiälä data spanned three years, containing more data points for model training, whereas the Värriö data only covered the period from April to August 2019, excluding the entire cold season when $H_2SO_4$ concentrations are significantly lower than those during the warm season (Jokinen et al., 2022). As a result, our model 3 (Fig. 3(a3), (b3), (c3)) and 4 (Fig. 3(a4), (b4), (c4)) can predict $J_5$ for boreal forest environment on a satisfactory level, including the possibility to use the estimated $H_2SO_4$ concentration from proxies as input. Nevertheless, limitations regarding precursor vapor production rate could potentially influence the prediction accuracy.

### 4.1.2 Urban-influenced: Beijing and Budapest

In anthropogenic emissions dominated region, such as Beijing, the measured and modelled $J_5$ are well aligned around the 1:1 line using model 3 (Fig. 3(d3), (e3), (f3)), and model 4 (Fig. 3 (d4), (e4), (f4)). In Beijing, a polluted megacity, the dominating precursor type has been found to be $H_2SO_4$–amine clusters (Cai et al., 2021). As expected, the testing result showed dramatic underestimations for Beijing using model 1 with only $H_2SO_4$ concentrations considered (Fig. 3(d1)), whereas models 2 (Fig. 3(d2)) and 3 (Fig. 3(d3)) yielded clearly enhanced $J_5$ predictions, with relatively minor differences between models 2 and 3. These features are consistent with the fact that in addition to $H_2SO_4$, also other vapors are import to NPF and sub-5 nm particle growth in Beijing, and demonstrate that RH and CS in our parameterization together determine in a complicated way the sources and sinks of these vapors, the survival probability of sub-5 nm particles, and the potential emissions of sub-10 nm primary particles from traffic. This study did not include amine-related compounds in the formulas because the lack of measured $NH_3$ data makes the parallel comparisons difficult among the chosen sites for model training.

For Budapest, a large European city, the underestimates in modelled $J_5$ are not as much improved as they were for Beijing when including RH or CS in the parameterization, which is indicative of distinct particle formation pathways between Beijing and Budapest, even though both sites represent urban background environments. On one hand, it is worth noting that including RH (model 2, Eq. 3) resulted in a decrease in the correlation coefficients between the measured and modelled $J_5$ in Budapest from 0.54 to 0.46 (Table 3). This suggests that the role of RH in the NPF process in Budapest is less significant than other chosen inputs, despite previous indications that high RH levels have a strong potential to suppress NPF during non-event days in Budapest (Salma et al., 2021), even though the RH values in Budapest were considerably higher than those in Beijing (Table S2). On the other hand, including CS (model 3, Eq. 4) in addition to RH (model 2, Eq. 3) leads to an increase in the correlation coefficients between the measured and modelled $J_5$ from 0.46 to 0.61 (Table 3). We used both NPF and non-NPF days during model training even though it was found that CS was 50% lower during non-NPF events in Budapest than the values during NPF events (Salma et al., 2016). As a result, it is difficult to determine if the model's performance gain was entirely brought on by the addition of CS. Otherwise, the results are all in line with the earlier indirect evidence that chemical species other than $H_2SO_4$ influence the particle growth and possibly NPF process in Budapest (Salma and Németh, 2019). If one considers additional vapors other than $H_2SO_4$ for Budapest alone for $J$ parameterization, one could include oxidation products of VOCs originating from either urban vegetation emissions or traffic emissions. For example, isoprene oxidation products can be used to describe the inhibiting effect on NPF (Heinritzi et al., 2020; Kiendler-Scharr et al., 2009), while monoterpene oxidation products could enhance sub-3 nm particle growth (Kulmala et al., 2013).

Based on the testing results, model 3 is more likely to predict a more accurate $J_5$ for Beijing based on the highest AIC ratio (except for 1), while model 1 predicts better for Budapest. Notice the fact that $J_5$ showed distinct levels of measured $J_5$ dependence with RH and CS in Beijing and Budapest ($J_5$ and RH: Beijing: $r = -0.21$, Budapest: $r = -0.1$; $J_5$ and CS: Beijing: $r = -0.02$, Budapest: $r = 0.57$, Fig. S1).

#### 4.1.3 Mediterranean rural site: Agia Marina, Cyprus

For Cyprus, it appears that meteorology and condensation sink terms have only minor effects on the formation of 5 nm particles in such rural environment under the influence of marine vapors when comparing the results from testing dataset across models (Fig. 3g). However, including RH improves slightly the correlation between the modelled and measured $J_5$ as seen in model 2 ($r$ rises from 0.42 to 0.49, Table 3). The reduced values of $r$ in model 3 (Eq. 4) indicate a somewhat reversed impact of CS on $J_5$, which requires additional examination as $J_5$ and CS are weakly correlated ($r$ = 0.03, Fig. S1). The $H_2SO_4$ concentration showed a low contribution to $J_5$ with the exponent being less than 1 in model 4 (Table 3), which led to a more underestimated modelled $J_5$ comparing to model 1 (Fig. 3(g1), (g4)). This could be an indication that potentially other anthropogenic, biogenic or marine compounds are of greater contribution to the particle formation processes in Cyprus than $H_2SO_4$ (Debevec et al., 2018). Owing to the orographic conditions, the air mass types approaching to the Cyprus measurement site are mixed, including the ones from North Africa, Marine, Europe, and northwest/southwest Asia. This results in the Mediterranean atmosphere in Agia Marina containing various vapors that could influence NPF. The potential key contributors could include oxidation products of dimethyl sulfide (DMS) originating from ocean plankton emissions (Rosati et al., 2021), iodine oxidation products like $HIO_3$ (He et al., 2021), the stabilizing agent $NH_3$ (Jiang and Xia, 2017; Lan et al., 2021; Lehtipalo et al., 2018; Yu et al., 2018), and oxidation products of VOCs from the surrounding pines forests and oaks under favorable meteorological conditions (Debevec et al., 2018).

We should note that the measurements in Cyprus covered only two weeks in springtime, which limited our quantitative observations in model training for other seasons compared to sites with long-term measurements. Based on the findings above, model 1 seems to be the most suitable functional form for the prediction of $J_5$ in Cyprus.

#### 4.1.4 Amazonian basin: Manacapuru, Brazil

The measured and modelled values of $J_5$ from the Manacapuru site scatter around the 1:1 line in all the models (Fig. 3). Previous studies reported high RH levels year-round in the measurement site near Manacapuru (Myers et al., 2022; Zhao et al., 2022), which is expected to suppress NPF frequency and to lead to lower formation rates. We observed such suppression effect when taking RH into account as shown by the increased correlation coefficients from 0.004 to 0.19 (Table 3). Studies from Manacapuru suggested that the epoxide vapors could be a potential precursor vapor in particle formation because of anthropogenic influences (Paulot et al., 2009), while Xu et al., (2014) suggested the presence of epoxide vapors can enhance particle nucleation when RH levels increase. We did not observe apparent improved model performance in model 3 when CS is included, as $r$ remained almost unchanged compared to model 2 (Table 3). One factor to consider is that we did not apply hygroscopic growth factor when calculating CS for Manacapuru to maintain the consistency of the training dataset. However, the impact of RH on CS, particularly on the actual particle surface area available for $H_2SO_4$ uptake, seems to be significant for high RH environments like Manacapuru (Myers et al., 2022). Another assumption could be that even with the high CS, it is still low enough to allow sufficient precursor vapors contributing to NPF processes.

These current findings provide evidence for $H_2SO_4$ being an effective enough precursor for the particle formation at 5 nm in the atmosphere of Manacapuru (model 1, Fig 3(h1)). However, the RH stabilization effect on $H_2SO_4$ is not exerted necessarily, as RH remains at high values at around 89 ±13 % despite whether it is measured during wet season or dry season (Myers et al., 2022). With these observations, model 1 with a focus on the $H_2SO_4$ concentrations manages to predict $J_5$ well for biogenic vapor dominated environment like Manacapuru.

## 5 Tracer model 5 simulation

We simulated the particle number size distribution (PNSD) in EC-Earth global chemical transport model TM5-MP (Tracer Model 5, Massively Parallel version, details in supplement) by applying it with our $J_5$ model 1 and 4. Together, we compared our simulation results with the acid-organic binary homogeneous nucleation model from Riccobono et al., (2014):

$$J_{\mathrm{Riccobono}} = k_{\mathrm{m}} \times [\mathrm{H_2SO_4}]^{\mathrm{p}} \times [\mathrm{BioOxOrg}]^{q} \quad , \qquad \text{(Eq. 8)}$$

where $k_{\mathrm{m}} = 3.27 \times 10^{-21}$ cm$^6$ s$^{-1}$, p = 2 and q = 1.

The details of the 14 tested measurement stations are shown in Table S7. Note that the data from these 14 stations are independent from any training or testing datasets used in the previous sections of this paper. Here, we essentially compared the simulated and measured PNSD in three particle modes (nucleation, Aitken, accumulation) from the entire year 2018 to assess the simulation accuracy among global environments.

Figure 4 shows the comparisons of PNSD between the on-site measurements and the TM5-MP simulations. For biogenic environments, simulations using model 4 shows the closest particle number size distribution to the measured ones particularly in Aitken mode particles, promoting the sulfuric acid-based nucleation mechanism involving the source-sink-meteorology even for environment dominated by biogenic vapors. For the Arctic region, model 4 simulated particle concentrations are overall overestimated, while model 1 simulation shows better alignment of particle number concentrations around the Aitken mode. This might indicate that the nucleation process has a lower dependence on the variations of meteorology than we expected. For coastal environments, even though Utö (Baltic Sea Island) and La Réunion (southern hemisphere island) are located at different hemispheres and also have different geographical settings, the nucleation mechanisms from models 1 and 4 both show similar predictions on particle concentrations across particle modes, with larger underestimation in the accumulation mode for model 1. This once again validates the source-sink-meteorology mechanism in model 4. By observing the ratio between the simulated and measured particle number concentrations, we can quickly see that the sulfuric acid-based particle formation mechanisms with (model 4) or without (model 1) meteorology inputs have successfully narrowed the gap between the simulations and observations across all particle modes, with significant improvements for the nucleation mode (Fig. 5). The "Total" contains the simulated/measured particle number concentrations ratio from all particle modes, and it is obviously seen that applying model 4 improves the overall global PNSD simulation compared with the sulfuric acid-organic vapor binary model from (Riccobono et al., 2014). This observation shows that including the RH and CS is needed for better understanding of the global particle number size distributions.

## 6 Conclusion

The particle formation rate is one of the key characteristics in new particle formation studies. By utilizing distinct field measurement data, we can model the particle formation rate and estimate the overall atmospheric aerosol budget over different environments. We parameterized $J_5$ in four functional forms using the combined datasets from six environments, covering boreal forests (Hyytiälä, Värriö), urban sites (a megacity of Beijing and a large European city of Budapest) and rural environment (Cyprus, Manacapuru). The particle formation schemes involve the main precursor vapor $H_2SO_4$, relative humidity (RH) and condensation sink (CS). Due to the small number of parameters and the diversity of environments included to generate the schemes, the roles of RH and CS are not only related to their potential direct impact on $J_5$, but also to sources and sinks of vapors other than $H_2SO_4$ contributing to formation and growth of sub-5 nm particles and to potential emissions of sub-10 nm particles, e.g., from traffic. Overall, our models showed improved performances as RH and CS were taken into consideration. The model evaluations may suggest that particle formation mechanism is more sensitive to certain factors in specific environments. Sulfuric acid is an effective precursor vapor in NPF processes for most of the measurement sites we selected for model training. Nevertheless, relying solely on $H_2SO_4$ generally resulted in a weaker model performance for environments where the NPF schemes are dominated by biogenic emissions. This suggests that for developing globally applicable particle formation rate models, more precursor vapor types need to be included alongside $H_2SO_4$.

The purpose of the paper is twofold: first, to address the lack of knowledge regarding global particle formation rates for particles at 5 nm and larger, and second, to provide a globally applicable semi-empirical parameterization for the sulfuric acid-based neutral particle formation. The simplicity of the parameterization is demonstrated by three factors. First, NPF is a widespread occurrence in various types of environments, where the characteristics of particle formation share common mechanisms involving major precursor types and environmental factors. Second, the main input $H_2SO_4$ concentrations data can be obtained from field measurements or proxies, from which the contribution of $H_2SO_4$ to NPF can be directly compared among global sites. Third, we skip the microphysics complexity of sub-5 nm particles, where the physical and chemical properties differ significantly from those that are above 5 nm when discussing particle formation and growth.

The limited data availability from certain sites (less than 1 year), such as Budapest, Cyprus and Manacapuru, should be noted when applying our models. Conclusions drawn from these sites can be more confidently applied to the specific seasons covered in the model training, such as springs being more representable for Budapest and Cyprus, and summer to early autumn for Manacapuru.

Overall, our parameterization findings show that our models including $H_2SO_4$ concentration, RH and CS can predict $J_5$ on a satisfactory level for various environment types at once. Among the tested models, models 3 and 4 (Eqs. 4 and 5) can be utilized for predicting $J_5$ in a global scale if (1) the $H_2SO_4$ concentrations are known whether through field measurement or proxies, (2) the meteorology parameter RH is monitored continuously, and (3) the particle number size distributions are sufficient and assessed to yield CS. Some caution should be maintained when utilizing these models for environments with very low RH and/or high CS, especially if the high CS is related to primary particle emissions, as the associations between these model parameters and $J_5$ are complicated and multifaceted. While the parameterizations presented in this study offer an improvement over previous approaches, further development is needed to incorporate vapors important for NPF, such as iodine oxoacids, particularly in marine environments.

*Author contributions.* Measurements: NS, RB, CY, LQ, TJ, IS, MV, TW. Data Analysis: XL, LD, MZ, NS, RB, CY, LQ, IS, PZ. Results interpretation: XL, LD, PP, TN. Discussions: all co-authors. Writing: XL, LD, PP, V-MK, TN. Comments and revisions: all co-authors.

*Data availability.* The datasets used in this study are now available on Zenodo: https://zenodo.org/records/15295592. The data for the 14 global measurement sites are from EBAS database (https://ebas-data.nilu.no/, last access 13.12.2024).

*Conflict of interests.* At least one of the (co-)authors is a member of the editorial board of Aerosol Research.

*Code availability.* The MATLAB code used for the parameterization training in this paper is available on Zenodo: https://zenodo.org/records/15295592.

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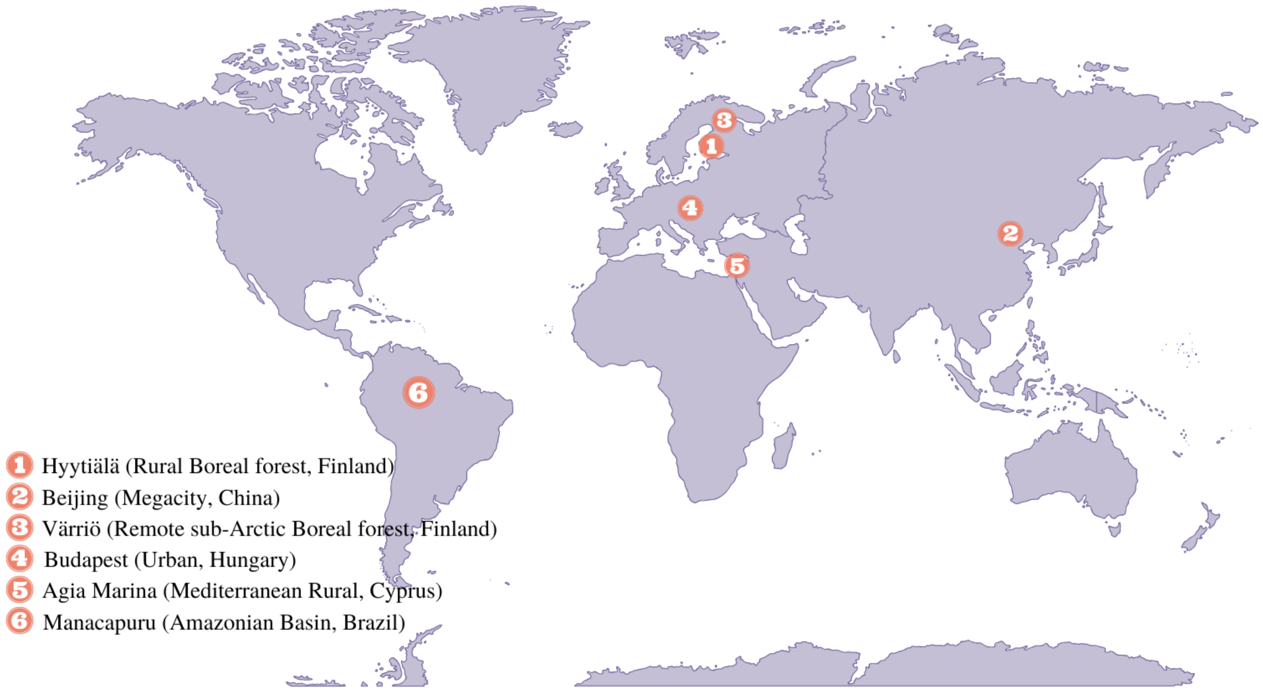


Figure 1. Map of measurement locations included in this study. The number markings indicate the exact locations of the measurements. Created using a template from Canva (www.canva.com)

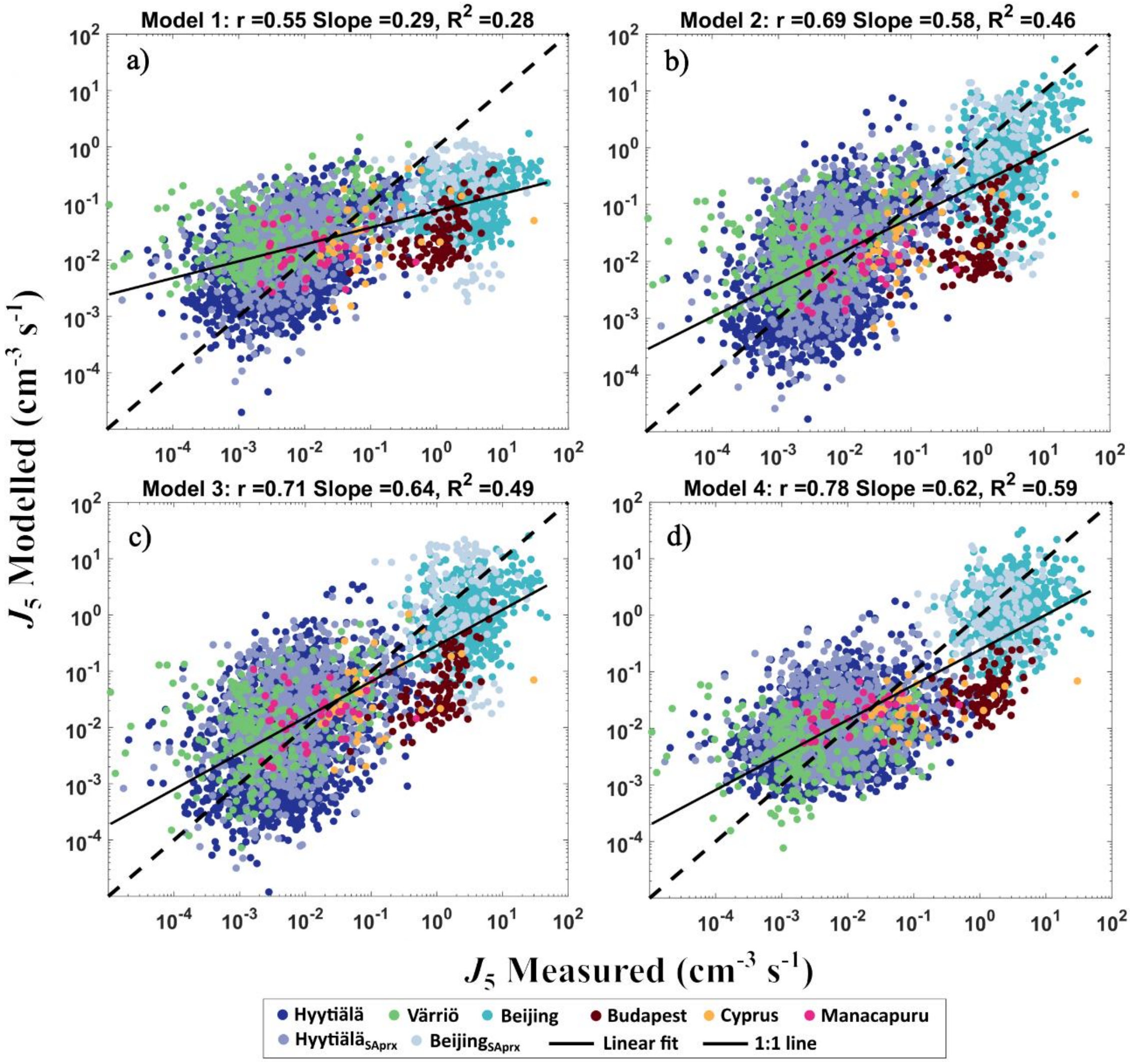


Figure 2. Modelled and measured $J_5$ scatterplots in logscale from four models using the testing dataset containing data from all sites in hourly time resolution. Each color represents the data from one measurement site, including datasets with $H_2SO_4$ proxy data from Hyytiälä and Beijing. The straight line showed the robust linear fit between the logscale modelled and the measure $J_5$ values, and the dashed line represented the 1:1 line. The correlation coefficient $r$, slope of linear fit, and the coefficient of determination $R^2$ are shown in the title of each subplot.

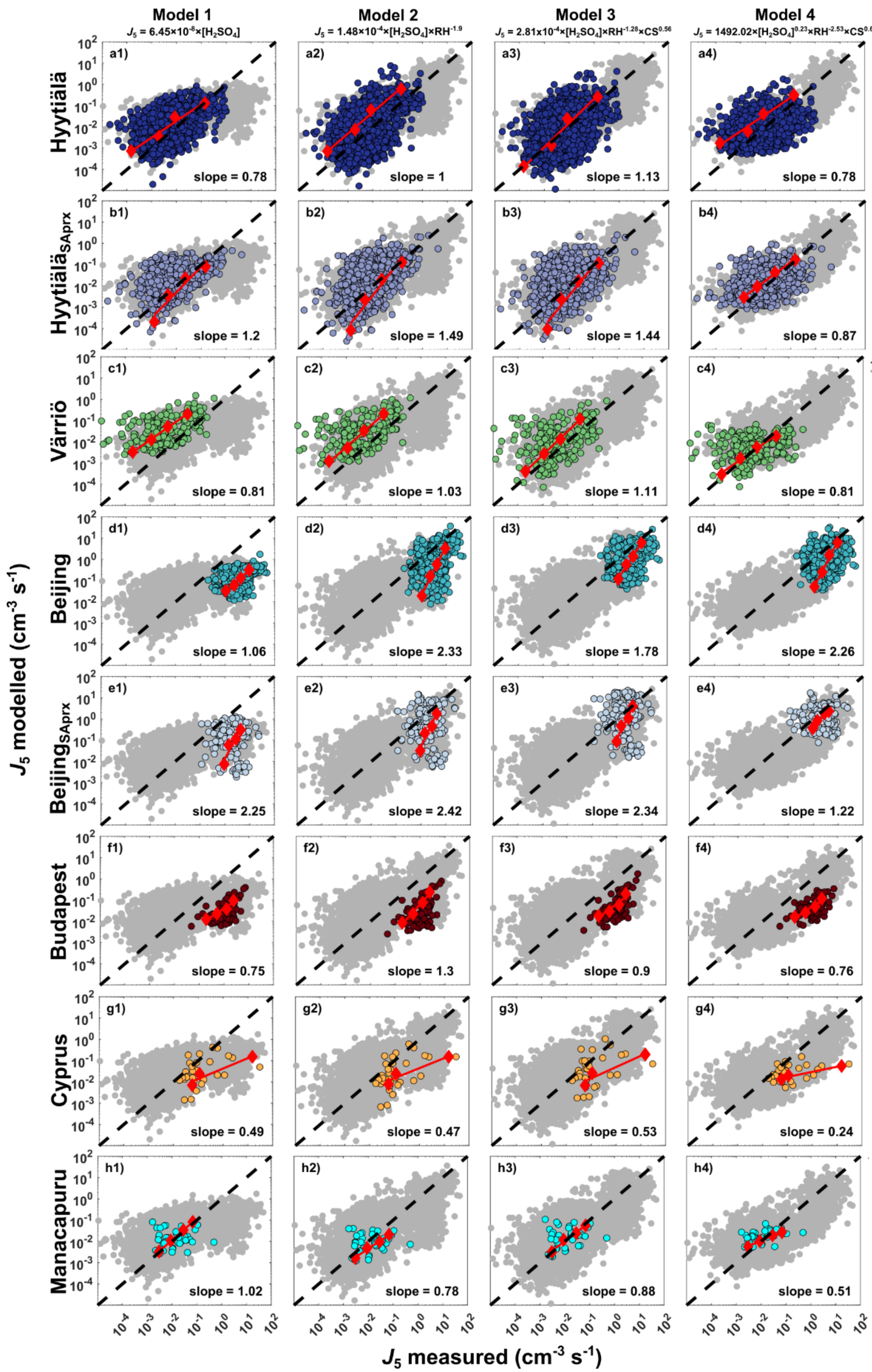


Figure 3. Modelled and measured $J_5$ scatterplots in logscale from four models using the testing dataset containing data from all sites in hourly time resolution. The labels on the left side of the y-axis are the site names. The

subscribed label "SAprx" indicates that the input $H_2SO_4$ concentrations was from $H_2SO_4$ proxies. The light grey scatters are all data points from the testing dataset, the colored scatters on top of them indicate the results from the corresponding measurement site. The red diamonds are the binned daily medians to show the temporal aggregation of the model performances on daily scale data. Overall, on a daily scale presents excellent performances on model 4 for boreal forest environment (a4, b4 and c4), polluted cities (d4, e4) and organic vapor dominated high humidity region (h1-h3). The red solid lines represent the linear fit on the binned hourly medians. The dashed line is the 1:1 line.

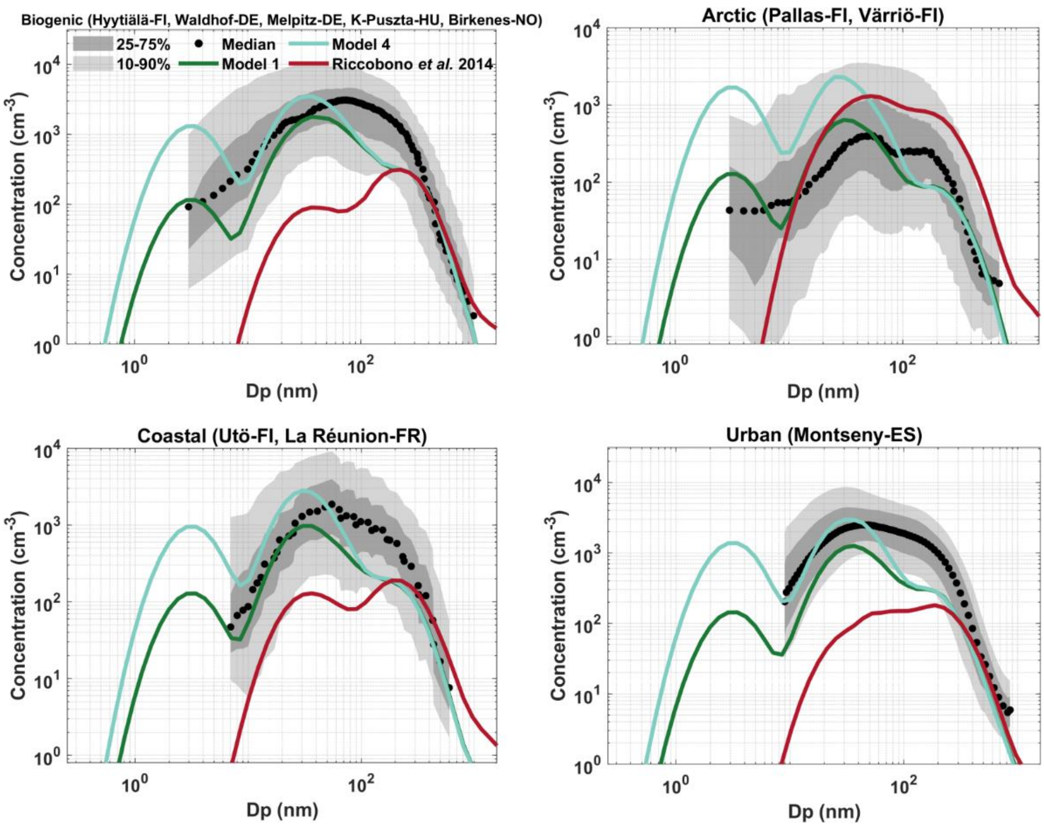


Figure 4. Environment-specific TM5-MP simulated particle number size distribution from 2018 in annual medians. Biogenic sites include rural and rural regional background environments (Hyytiälä, Waldhof, Melpitz, K-Puszta and Birkenes); Coastal sites cover islands on the Baltic Sea and on the Indian Ocean in the southern hemisphere close to Madagascar (Utö and La Réunion); Arctic sites are two Finnish sites both situate within the Arctic Circle (Pallas and Värriö); Urban site is represented by a Spanish city Montseny.

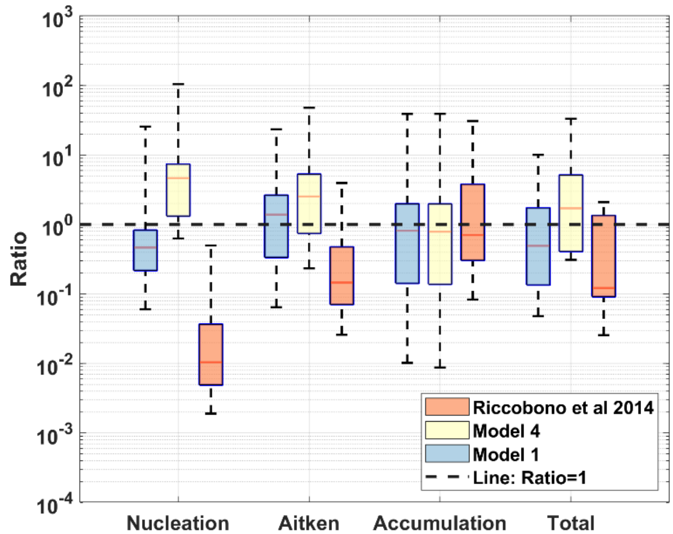


Figure 5. Ratio of simulated and measured particle number concentrations from the 14 global sites from TM5-MP simulations under three nucleation settings (Riccobono, model 1, and model 4) resulting in three particle modes (nucleation, Aitken, accumulation) in 2018 annual medians. The black line represents "ratio = 1" as a reference line. The "Total" represents the overall ratio between the simulation and the measurement particle number concentrations from all modes.

Table 1. Number of data points from each measurement site. The numbers in column "Total" account for the data points from six-site combined dataset utilized in model training and testing. The training set contains 75% of the total training data points, and 25% for testing set.

| Sites | Hyytiälä | Beijing | Värriö | Budapest | Cyprus | Manacapuru | $\text{Hyytiälä}_{proxy}$ | $\text{Beijing}_{proxy}$ | Total |
|---|---|---|---|---|---|---|---|---|---|
| Training | 5003 | 1342 | 728 | 367 | 140 | 140 | - | - | 7720 |
| Testing | 1642 | 501 | 248 | 109 | 34 | 40 | 797 | 164 | 3535 |

Table 2. Coefficient values ($k_{\mathbf{x}}$, $k_{RH}$, $k_{CS}$, $k_{SA}$) retrieved from parameterization using training dataset. The term *SSe* represents the sum of squared error of each model. The units of $k_{\mathbf{x}}$ ($x$ = 1, 2, 3, 4) vary as the functional form of model changes, while $k_{RH}$, $k_{CS}$, $k_{SA}$ do not contain units. Since RH is counted using percentage (%), a dimensionless number, the scaling coefficients k0 count mainly the units from $H_2SO_4$ concentrations and CS. As such, we must ensure that the RH input is in percentage.

| Models | Functional forms | $k_x$ | $k_{RH}$ | $k_{CS}$ | $k_{SA}$ | *SSe* |
|---|---|---|---|---|---|---|
| 1 | $k_1 \times [H_2SO_4]$ | 6.45E−8 ($s^{-1}$) | | | | 2.78E+03 |
| 2 | $k_2 \times [H_2SO_4] \times RH^{k_{RH}}$ | 1.48E−4 ($s^{-1}$) | −1.9 | | | 5.16E+03 |
| 3 | $k_3 \times [H_2SO_4] \times RH^{k_{RH}} \times CS^{k_{CS}}$ | 2.81E−4 ($[s^{-1}]^{0.45}$) | −1.28 | 0.56 | | 5.18E+03 |
| 4 | $k_4 \times [H_2SO_4]^{k_{SA}} \times RH^{k_{RH}} \times CS^{k_{CS}}$ | 1492.02 ($[cm^{-3}]^{0.78} \times [s^{-1}]^{0.33}$) | −2.53 | 0.67 | 0.23 | 3.36E+03 |

Table 3. Summary of overall and site-specific correlation coefficients (r) four models using the testing dataset. The numbers in brackets under the site name**s** represent the count for data points.

| | | Slopes and *r* (robust linear fit), logscale | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | Models | Hyytiälä (1642) | Beijing (501) | Värriö (248) | Budapest (109) | Cyprus (34) | Manacapuru (40) | $\text{Hyytiälä}_{SA}$ (797) | $\text{Beijing}_{SA}$ (164) | Overall |
| Slope | 1 | 0.43 | 0.33 | 0.32 | 0.58 | 0.35 | 0.04 | 0.34 | 0.07 | 0.30 |
| | 2 | 0.62 | 0.57 | 0.40 | 0.66 | 0.48 | 0.15 | 0.57 | 0.23 | 0.58 |
| | 3 | 0.48 | 0.43 | 0.32 | 0.85 | 0.37 | 0.18 | 0.42 | 0.12 | 0.64 |
| | 4 | 0.25 | 0.28 | 0.12 | 0.48 | 0.17 | 0.24 | 0.28 | 0.12 | 0.62 |
| *r* | 1 | 0.43 | 0.30 | 0.44 | 0.54 | 0.42 | 0.04 | 0.41 | 0.004 | 0.55 |
| | 2 | 0.47 | 0.32 | 0.47 | 0.46 | 0.49 | 0.19 | 0.48 | 0.07 | 0.69 |
| | 3 | 0.37 | 0.30 | 0.35 | 0.61 | 0.38 | 0.21 | 0.36 | 0.02 | 0.71 |
| | 4 | 0.31 | 0.22 | 0.18 | 0.51 | 0.37 | 0.46 | 0.33 | 0.09 | 0.78 |