Answers to the Second Reviewer's Comments

Parameterization of particle formation rates in distinct atmospheric environments

Xinyang Li et al., 2025

The line numbers in bold refer to indicating the changes made in the manuscript to be further uploaded.

1. Sulfuric acid proxy data, and in line 225. The reference from Dada is provided, but are the calculations shown for these datasets mentioned anywhere in the manuscript or supplementary material?

   Reply:

   We thank the reviewer for the thoughtfulness about the sulfuric acid proxy details. The proxies are easily accessible in Dada et al., (2020), with clear labels for Hyytiälä and Beijing including the values of each coefficient (Table 1, Eq. 9 and 12). We can provide the proxy data upon request.

2. Line 218. What would be the effect of considering the correction for hygroscopic growth?

   Reply:

   We appreciate the reviewer for the insight into the basics of hygroscopic growth particles. When we consider hygroscopic growth, the particle sizes will increase by absorbing water from the ambient environment, which is dependent on the relative humidity level. As these pre-existing particles grow, the likelihood of condensable vapors losing onto their surfaces increases. These speculations are based on the hygroscopicity of the pristine boreal environment of Hyytiälä. When we apply the Hyytiälä parameterization to other environments where the hygroscopicity varies differently, it may cause over estimation or underestimation of the CS and CoagS.

   We will add the following text in section 2.3.3 (L218) to support the decision of excluding the hygroscopic correction on CS and CoagS.

   Additional text in L218-222:

   "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 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."

Furthermore, since we have RH in our parameterization combined with CS without a hygroscopicity correction, the increased sink due to hygroscopic growth of particle is transferred from CS to RH in our parameterization. We modified the text in section 3.1 into the following form (**L260-269**):
"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 with 1) either 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."

3. Line 222. The detection limit of H_2SO_4 is exceptionally good at 5×10^3 molecules per cubic centimeter. Was this case for all the mass spectrometers considered here?

   Reply:

   We thank the reviewer for highlighting the H_2SO_4 limits. Yes, this threshold is applied to all the H_2SO_4 data collected from the six sites using mass spectrometers. We are

aware that there are numerous NPF pathways involving H_2SO_4 that can compete with the mechanism based solely on H_2SO_4, therefore, we set a limit to ensure that avoid the artifact inaccuracy in the measurements, while also maintaining a high enough H_2SO_4 concentration level to pronounce the particle formation pathway based on H_2SO_4. Although the uncertainty in H_2SO_4 observations below 10^5 molecules cm^{-3} are already large, it is still useful to continue the parametrizations towards small concentrations, since it is better to have a suitable parametrization which is consistent with higher concentrations than no parametrization at all in the low concentration range.

4. Can the authors clarify how low RH and NPF are related? It is not entirely clear to me. Similarly, I do not fully understand how this phenomenon can be seen on Figure 3 as indicated in lines 355-357.

   Reply:

   We thank the reviewer for the concern on the statement about RH. In section 3.1.1, we mentioned the relation between global radiation and RH, and we have adapted the comments also from reviewer 1 that RH and NPF have correlations but not necessarily causation. We will modify the text accordingly to resolve the unclarity in the statement:

   Current text in the current manuscript, from L355-357:

   "which confirmed the strong relationship of the low RH favoring NPF occurrence for the boreal forest environmental conditions (Dada et al., 2018; Hamed et al., 2011)"

   Text modified from L355-357 **(Now L383-386)**:

   "which demonstrates a potential NPF favoring condition associated with increased global radiation for the boreal forest environment (Dada et al., 2018; Hamed et al., 2011), as well as lower sink for vapors and growing sub-5 nm particles (see section 3.1), under low RH conditions".

5. I understand the lack of data availability for HOMs and NH_3. However, when describing NPF in polluted environments, NH_3 is essential. Do you think an NH_3 proxy can be used in the future? How might the results change if NH_3 were included? Could you also comment on how the addition of ammonia will influence the model, and especially which coefficients will be influenced by that and why? I would be very interested in hearing your thoughts on this.

   Reply:

We gratefully appreciate the reviewer for the critical insight about considering NH_3 into the future parameterization. Though NH_3 cannot enhance NPF alone, it is rather responsible for stabilizing H_2SO_4 in combinations with other organic vapors to enhance NPF. It will be a great compliment if NH_3 can be added in future parameterizations. We provided reliable examples of applying the H_2SO_4 proxy in our models, therefore, we believe an NH_3 proxy can also be implemented if they are able to capture the NPF pathway as precisely as possible and are well-validated. The NH_3 proxies will need to be first tested on specific environmental conditions (i.e. the correlations between NH_3 and meteorology, sinks, vapor source rate, boundary layer height, etc.), then generalized reasonably for global simulations.

Lehtipalo et al., (2018) showed that the addition of NH_3 can efficiently enhance particle formation by stabilizing H_2SO_4 to interact with HOM. Hence, we expect to see an overall addressed underestimation of J_5 by adding NH_3 into the models, particularly in boreal forests (Hyytiälä, Värriö), where HOM level is high for forming H_2SO_4-NH_3-HOM clusters during nucleation processes, and in rural environments (Agia Marina in Cyprus, Manacapuru in Brazil), where there are possible transported NH_3 from agriculture lands.

Concerning the coefficients, we must analyze the correlations between NH_3 and RH, CS, and H_2SO_4 in all environments. Consider the extremely clean (Hyytiälä) and extremely polluted environments (Beijing) as examples, NH_3 is potentially positively correlated with temperature (T) in Hyytiälä springtime due to the intensified gas released from the ground during the melting season (Hemmilä et al., 2018), which could also be interpreted as NH_3 being negatively correlated with RH, because T and RH are anti-correlated in Hyytiälä. The rising T is related to rising solar radiation, which would accelerate the water evaporation in the atmosphere and reduces RH. In Beijing, a study about the NH_3 behavior in urban and suburban revealed that the correlations between NH_3 and the ambient water mixing ratio vary seasonally. However, an overall negative correlation could be seen (Fig. 4, Lan et al., 2021). If applying these findings to our J_5 models, the coefficient on RH should be lower particularly for spring data after adding NH_3, thereby an overall smaller k_{RH}.

6. Lines 249-250. Please add more references, particularly when discussing not only ammonia but also HNO_3. The authors could mention, for example, the synergetic NPF mechanism including sulfuric acid + ammonia + nitric acid.

Reply:

We thank the reviewer for providing sharp focus on enriching the statement about the significant role of NH_3 involving HNO_3. We modify the text as follows:

Current text in the current manuscript, from L248-251:

"As for precursor vapor types other than sulfuric acid, although the 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), we were unable to include these vapor concentrations owing to limited data availability from the chosen measurement sites."

Text modified from L248-251 (now **L273-277**):

"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."

7. Lines 319-321. What do the authors think could be a potential mechanism for NPF in Budapest during spring, as supported by results described in Section 4.1.2?

Reply:

We thank the reviewer for bringing the NPF mechanism in the urban site in Budapest to our attention. When we solely focus on Budapest, even though we considered only the H_2SO_4-driven nucleation, the VOCs from the urban vegetation and the traffic emissions are likely relevant in the local NPF (Kulmala et al., 2022). The BVOCs in urban environment can be related to isoprene and monoterpene, where isoprene shows inhibiting effect on NPF (Heinritzi et al., 2020; Kiendler-Scharr et al., 2009), while the low volatile oxidation products from monoterpene could enhance sub-3 nm particle growth (Kulmala et al., 2013).

The most commonly discussed traffic emissions include SO_2 and NOx (Brean et al., 2024), with NH_3 and HNO_3 also influencing urban NPF under certain meteorological conditions (Jiang and Xia, 2017). Based on these findings, we speculate the NPF mechanism in Budapest primarily involves H_2SO_4 condensation (potentially stabilized by NH_3), the ternary nucleation pathway of HNO_3-H_2SO_4-NH_3 (RH at ~60%, which is particularly the median RH in our study), oxidation of VOCs from urban vegetations and combustion for particle growth on larger sizes, and finally the suppressing effect of NOx.

We will add the following supporting text in Section 4.1.4 based on the statements above.

Additional text from L392 **(Now L422)**:

"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)."

8. Lines 398-400. Which marine vapors could improve the performance of the model? Additionally, in lines 405-406, could the author indicate which vapors are expected to influence NPF, along with relevant references?

   Reply:

   We appreciate the reviewer's valuable insight about the details in Cyprus NPF mechanism. The atmosphere in the Mediterranean Sea in Cyprus is mixed with various sources of vapors that can contribute to the local and regional NPF. Firstly, we expect to include dimethyl sulfide (DMS) from the oxidation of sulfur emissions from plankton in the ocean (Rosati et al., 2021). Secondly, iodine oxidation products such as HIO_3, which the marine NPF is sensitive to, should be considered (He et al., 2021).

   For other potential vapors for NPF contribution, we would like to focus on NH_3 from the local or transported sources as they have stabilizing effect on H_2SO_4 to enhance NPF (Jiang and Xia, 2017; Lan et al., 2021; Lehtipalo et al., 2018; Yu et al., 2018). Moreover, oxidation products of VOCs from the surrounding biogenic sources (pines forests and oaks) originated from monoterpene and isoprene emissions also contribute to NPF in Agia Marina with favorable meteorological conditions (Debevec et al., 2018).

   We will add the following supporting text in Section 4.1.4 based on the statements above.

   Additional text from L408 **(Now L442-447)**:

   "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)."

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