Answers to Referee Comments

Parameterization of particle formation rates in distinct atmospheric environments

Xinyang Li et al., 2025

Part 1: List of all changes made in the manuscript following the referee comments

1. Modification for RC1, Comment 4,5,6 combined. Part 5/6. **(L24-26)**
   "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."

2. Additional text considering the overall Referee Comments. **(L204-208)**
   "Ideally, this term, as well as the concentration of particles within the size range, Ndp, 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)."

3. Additional text for RC1, Comment 1. Part 1/2. **(L224-236)**
   "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."

4. Additional text for RC1, Comment 1. Part 2/2. Also for RC2, Comment 2. **(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 H2SO4 as an input variable, the evidently important sink effect of a pre-existing particle population on the ambient H2SO4 concentration is implicitly transferred from CS to H2SO4 in our parameterization. Indirectly, CS may also be associated, either causally or not, with 1) emissions or sinks of vapors other than H2SO4 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."

5. Modification for RC2, Comment 6. **(273-277)**
***"**Other than sulfuric acid, highly oxygenated organic molecules (HOMs) and ammonia (NH3) 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 H2SO4-NH3-H2O (Yu et al., 2018) and HNO3-related clusters such as HNO3-H2SO4-NH3 in the upper tropospheric particle nucleation (Wang et al., 2020, 2022, 2023). Yet, we are unable to include HOM nor NH3 concentrations owing to limited data availability from the chosen measurement sites."*

6. Additional text considering the overall Referee Comments. **(L294-297)**
"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 H2SO4 participating in particle formation and growth and, in polluted environments, to sub-10 nm particle emissions from traffic."

7. Modification for RC1, Comment 4,5,6 combined. Part 2/6. **(L377-379)**
"It is notable that with the combined data sets, the condensation sink receives a positive exponent in models 3 and 4 (kCS = 0.56 and 0.67, respectively), likely due to its association with concentrations of other condensable vapors than H2SO4 and traffic emissions."

8. Modification for RC1, Comment 3. Also for RC2, Comment 4. **(L383-386)**
"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)."

9. Modification for RC1, Comment 4,5,6 combined. Part 1/6. **(L399-405)**
"As expected, the testing result showed dramatic underestimations for Beijing using model 1 with only H2SO4 concentrations considered (Fig. 3(d1)), whereas models 2 (Fig. 3(d2)) and 3 (Fig. 3(d3)) yielded clearly enhanced J5 predictions, with relatively minor differences between models 2 and 3. These features are consistent with the fact that in addition to H2SO4, 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."

10. Addition text for RC2, Comment 7. **(L422-426)**
"If one considers additional vapors other than H2SO4 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)."

11. Addition text for RC2, Comment 8. **(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 HIO3 (He et al., 2021), the stabilizing agent NH3 (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)."

12. Modification for RC1, Comment 8. **(L466-470)**
"These current findings provide evidence for H2SO4 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 H2SO4 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 H2SO4 concentrations manages to predict J5 well for biogenic vapor dominated environment like Manacapuru."

13. Modification for RC1, Comment 4,5,6 combined. Part 3/6. **(L506-511)**
    "The particle formation schemes involve the main precursor vapor H2SO4, 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 J5, but also to sources and sinks of vapors other than H2SO4 contributing to formation and growth of sub-5 nm particles and to potential emissions of sub-10 nm particles, e.g., from traffic."

14. Modification for RC1, Comment 4,5,6 combined. Part 6/6. **(L531-532)**
    "Overall, our parameterization findings show that our models including H2SO4 concentration, RH and CS can predict J5 on a satisfactory level for various environment types at once."

15. Modification for RC1, Comment 4,5,6 combined. Part 4/6. **(L535-537)**
    "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 J5 are complicated and multifaceted."

16. Modification for RC1, Comment 9. **(L538-540)**
    "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."

Part 2: Combined replies to the comments from Referee 1 and 2.

Answers to the First Reviewer's Comments

1) L217-218: I think the correction for hygroscopic growth was necessary for the intercomparisons. The CS and CoagS terms may vary substantially depending on the RH and the aerosol chemical compositions.

Reply:

We appreciate the reviewer highlighting the concern of the hygroscopic correction on CS and CoagS. The goal of not implementing the hygroscopic correction is mainly to harmonize the data composition and the later model analysis.

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

Additional text from L218 **(Now L224-236)**:

"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."

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 in L242 **(Now 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."

2) L248: It is understandable not to include HOMs and NH_3 in the parameterization due to the lack of data. However, it is more important to know how the exclusion of these compounds would impact the performance of these models. How sensitive are these models to the H_2SO_4 data, especially when H_2SO_4 proxies are used, which may lead to substantial uncertainty in H_2SO_4 input data?

Reply:

We agree that adding HOM and NH_3 could potentially enhance the model performance further. For HOM, our first consideration was that the definition of HOM varies from environment to environment depending on the primary emission types and oxidizing compounds. It leads to challenges in generalizing the molecular mass range of HOM for global model parameterization. Here we can provide the comparison result of model trainings with and without adding HOM data (same measurement period as H_2SO_4) from Hyytiälä. From Figure 1 below it can be seen that HOM is not improving the model, but quite the opposite as seen from the decreased Pearson *r* values. Therefore, we stayed with the most significant vapor H_2SO_4.

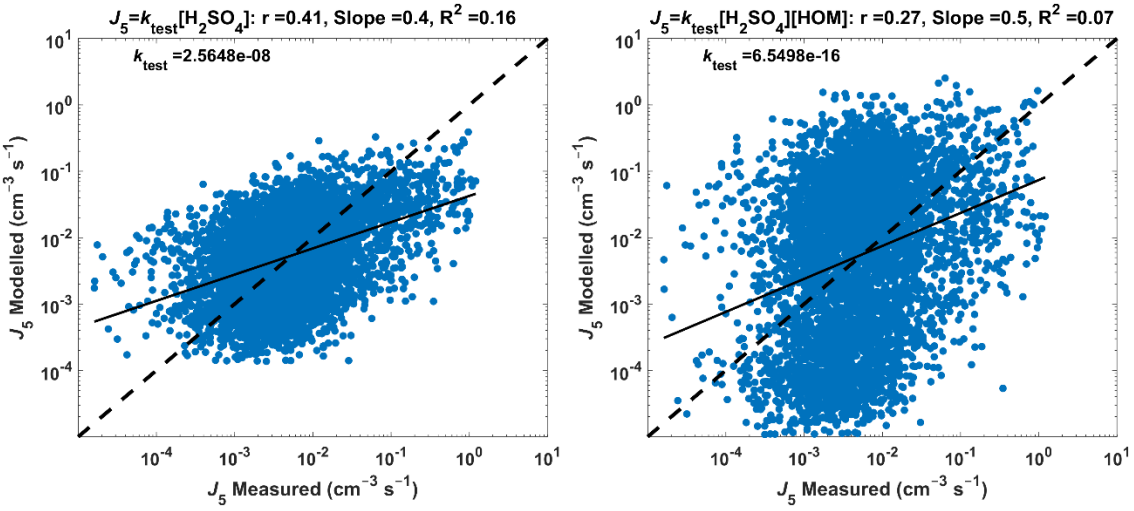

Figure 1. Model training with the functioning forms of $J_5 = k_{test}[H_2SO_4]$ (left), and $J_5 = k_{test}[H_2SO_4][HOM]$ (right) using Hyytiälä data.

For NH_3, it is indeed a very significant component in particle nucleation processes in many environments. Though we cannot provide test examples due to the unavailable NH_3 data in any of the chosen sites, we will explain our concerns here. NH_3 is emitted primarily from agriculture lands. In our case, the only agriculture-relevant environment is Agia Marina in Cyprus, where the vapor types are typically mixed among H_2SO_4, organic vapors, NH_3 and marine vapors. In addition, Cyprus data is short-term compared to other sites included (except for Budapest) in this study. Therefore, we speculate the effect of NH_3 is minor in our model training.

Both H_2SO_4 models for Hyytiälä and Beijing in Dada et al., (2020) have their slopes extremely close to 1 on testing dataset, which granted advantages to the models to be applied on a broader scale. According to our testing results (Table 3), the Pearson *r* differs little using the measured H_2SO_4 or H_2SO_4 proxy data in Hyytiälä. The difference in Beijing Pearson *r* between using the measured and proxy H_2SO_4 is possibly due to the data availability in the testing dataset. The measured H_2SO_4 in Beijing testing dataset ranges from May 2018 to April 2019 (almost one year), while the proxy data covers March to April 2019 (only in spring).

3) L356: Strictly speaking, NPFs were often associated with Low RH conditions, which does not necessarily mean that low RH favors NPFs. These two phenomena may concur due to the same underlying cause. For instance, stronger solar irradiation can lead to higher ambient temperature and, thus, lower RH. However, the real factor in intensifying NPF could be the increased atmospheric photooxidation capacity that led to more production of NPF gas precursors.

Reply:

In section 3.1.1, we mentioned the relation between global radiation and RH, and we agree with your suggestion and will modify the text accordingly:

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 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)."

4) L357: It is a bit strange that the higher SA and lower CS in Värriö would lead to a lower frequency of NPF.

5) L373: There is still no direct evidence that meteorology will significantly impact NPF in Beijing. The CS term was indeed playing a critical role in regulating NPF in Beijing.

6) L374-375: How did background aerosols sustain NPF? Did not the loss to preexisting aerosols compete with the formation of sub-5 nm particles? The high levels of background SO_2, VOCs, and their oxidation products may be responsible for the intense NPFs in Beijing.

Reply to comments 4, 5 and 6 together:

The lack of other stabilizing agents in Värriö (e.g. NH_3, amines, VOCs, etc.) may result in the low contribution of H_2SO_4 even when H_2SO_4 concentration is higher than that in Hyytiälä. For that, we would need measurement of the stabilizing gases to investigate further. Another speculation of the low NPF frequency in Värriö is that the organic vapor pathway may compete with the H_2SO_4 pathway at 5 nm particles, leaving high H_2SO_4 concentration seemingly less effective. As a result, we believe the statements given above are potentially the reasons for such NPF phenomena in Värriö. We will add the modifications in the manuscript accordingly.

We sincerely thank the reviewer for the critical and important comment on the meteorological impact on NPF in Beijing and appreciate that the reviewer pointed out an imprecisely addressed statement on CS in the text.

We have modified the text in the current manuscript in L370 **(Now L399-405)** into the following form:

"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."

We also added clarifications to Sect. 4.1 in L349 **(Now L377-379)**:

"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."

...in the conclusions in L465 **(Now L506-511)**:

"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."

...and additional texts in L490 **(Now L535-537)**:

"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."

...and in the abstract **(L24-26)**:

"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."

Additionally, we have replaced the wordings possibly interpreted to mean (direct) impacts of meteorology and vapor loss with expressions more clearly mentioning relative humidity and condensation sink as model parameters (**L466**).

7) L411: These results strongly suggested that precursors other than H_2SO_4 should be considered for these models to work appropriately in marine environments.

Reply:

We appreciate the reviewer for the insightful comment on the precursors in regards of marine environment in the parameterization. Indeed, iodine-driven nucleation can be more responsible than H_2SO_4-driven nucleation in marine environment during NPF events. However, iodine related vapors are confined within marine environment (Sipilä et al., 2010). When we do the parameterization for a single environment, we will consider thoroughly the key components in the local NPF characteristics. When parameterizing global environments, we aim for the models to be as representative and simple as possible to cover the most common NPF mechanism for the majority of environments. In addition, SO_2 is also transported globally and oxidized in the atmosphere above the marine regions, providing that its role as a precursor vapor in

marine NPF mechanisms cannot be neglected. Naturally, we strongly recommend conducting the J parameterizations in marine environments following the environment-specific NPF mechanism.

8) L426: The Manacapuru case may be very special. The RH was very high year-round, and thus, J_5 became insensitive to variations in RH and the corresponding aerosol hygroscopic growth, which may be treated as a constant. This may explain the better slope (1.02) found in model 1 simulation (Fig. 3h1).

Reply:

We would like to thank the reviewer for clarifying the RH impact on the NPF mechanism in Manacapuru. Manacapuru is a very interesting environment for our parameterization precisely because of the near insensitivity of RH on J_5 as compared to other locations. This is an observation acknowledging that RH likely doesn't play a dominant role in controlling NPF events in Manacapuru.

We will modify the text in L426-429:

"These current findings provide evidence for H_2SO_4 being an effective enough precursor for the particle formation at 5 nm, as well as the RH stabilization effect on H_2SO_4in the atmosphere of Manacapuru. So far, model 4 with a focus on the H_2SO_4 concentrations along with meteorology and vapor loss impacts manages to predict J_5 for biogenic vapor dominated environment like Manacapuru."

Text modified in L426-429 (**now L466-470**):
"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."

9) These J-models were developed to predict NPF rates globally, but they did not consider nucleation mechanisms involving iodine oxoacids (IO). Since ~70% of the Earth's surface is seawater, how would this affect the application of these J-models by omitting IO-related NPF mechanisms?

Reply:

We sincerely thank the reviewer for the critical comment on the choice of precursor in our global parameterization models. We believe part of our answers in point 7 can explain this question as well. Besides the global circulation of SO_2 and their oxidation

product H_2SO_4 over the marine regions, iodine products play a crucial role in marine NPF events. The iodine measurements take place usually at certain marine site, while they are usually not measured for inland regions, such as Hyytiälä, Värriö, Budapest, etc. It brings difficulties to obtain measured iodine oxidation products for the chosen locations in this study. However, we would strongly recommend including iodine related products when conducting *J* parameterization specifically for marine regions.

We add the following text to support our statement at L490 (**now L538**):

"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.".

Answers to the Second Reviewer's Comments

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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