We are very grateful to the evaluations from the reviewers, which have allowed us to clarify and improve the manuscript. Below we addressed the reviewer comments, with the reviewer comments in black and our response in blue or green.

Suggestions for revision from Anonymous referee #2:

The authors responded thoroughly to the review comments. The paper is good and should be highly cited. The scientific content is ready for publication, but ALL of the modifications to the paper need review for written English.

Response: We appreciate the positive feedback and note that the scientific content is ready for publication. However, we will ensure that all modifications undergo a thorough review for written English.

The summary and discussion could do with a real conclusion. It is currently a brief summary followed by a long discussion of a list of limitations of the study in a fairly random order (HOMs, then nucleation, then (anthropogenic) HOMs again). This responds to the review comments, but the structure does not really help the readability of the paper or facilitate scientific interpretation of the results. What are the most important uncertainties?

Response: Thanks for your insightful feedback regarding the structure of the summary and discussion sections. We have reorganized the sequence of discussions on the uncertainties in this study (Lines 638 to 774). The new order begins with a discussion on uncertainties related to the concentrations of nucleating species, including biogenic HOMs, anthropogenic HOMs and inorganic compounds, followed by the uncertainties in the NPF parameterizations. We have also highlighted that the greatest uncertainty arises from the concentration of biogenic HOMs (Line 638), which is attributed to uncertainties in the chemical reactions. The revised **Summary** is shown below. The Original text is in **Blue**, newly added text is **<u>Underlined</u>**, and the reordered paragraphs are in **Green**.

"<u>The chemical mechanisms of biogenic HOMs used in this study are state-of-the-art, but the largest uncertainties still come from the chemical mechanisms of HOMs and thus HOMs concentration.</u> Although we only consider two-step autoxidation reactions which are not the most advanced (Heinritzi et al., 2020; Simon et al., 2020), this impact on organic nucleation rate is almost negligible. Specifically, the number of autoxidation steps has almost no effect on the rate of heteromolecular nucleation of sulfuric acid and organics (HET), which is the most significant contributor to organic nucleation rate (Fig. 6 in the main text). This is mainly because the number of autoxidation steps affects neither the yield nor the concentration of C10-HOMs, only their molecular formulas and volatility. In our simulation, the lower volatility of C10-HOMs does not affect their participation in HET (i.e. LVOC, ELVOC and ULVOC can all contribute to HET), so the rate of HET is not influenced by the number of autoxidation steps. Previous studies (Kurtén et al., 2016; Tröstl et al., 2016) have already indicated that C10 class molecules alone do not have low enough vapor pressure to

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initiate the nucleation, without the presence of other species such as sulfuric acid or bases. This is further supported by the fact that C20 class molecules are mainly responsible for pure biogenic nucleation (Heinritzi et al., 2020; Frege et al., 2018). This means that C10-HOMs might become less volatile when undergoing one additional autoxidation step, transitioning from LVOC ($3 \times 10^{-5} < C^*(T) < 0.3$ µg m^{-3}, where $C^*(T)$ is the effective saturation concentration) to ELVOC ($3 \times 10^{-9} < C^*(T) < 3 \times 10^{-5}$ µg m^{-3}), but this is unlikely to affect the pure organic nucleation rate.

There might be some overestimations with C15 and C20 involved in new particle formation if we assume that all the accretion products are ELVOC or ULVOC. In the updated model, $C_{15}H_{18}O_9$ (C15, extremely low volatility) and $C_{20}H_{32}O_8$ (C20, ultra-low volatility) are just simplified representatives of all C15 and C20 dimers. While more dimer species with low volatility have already been detected on chamber experiments (Stolzenburg et al., 2018; Ye et al., 2019; Schervish and Donahue, 2020), they did not provide the explicit chemical kinetics of related reactions (i.e. the intermediate products and their yields) so it is hard to incorporate these reactions and species in the model. On the other hand, although yields of accretion products vary by 1 to 2 orders of magnitude in previous studies (Rissanen et al., 2015; Berndt et al., 2018; Zhao et al., 2018), the yields of C15 and C20 currently use are very low (4%), resulting in relatively low dimer concentrations. Even if they were all ELVOC and ULVOC, it would not lead to a significant overestimation, and therefore, would not substantially impact nucleation and growth rates.

Neglecting the oligomerization and decomposition of accretion products may affect their concentrations; however, these effects are negligible. Not accounting for the oligomerization can lead to higher volatility of aerosols, resulting in a reduction of the mass concentration in the particle phase and condensation sink (CS), but increased mass in the gaseous phase. This could lead to an overestimation of the NPF rate. Since the mass of HOMs-SOA accounts for only about 10% of the total SOA mass, the impact on NPF rate can be neglected. Not considering decomposition may result in an overestimation of the mass and number concentration of HOMs in the particle phase, and consequently an overestimation of CS and an underestimation of the NPF rate. However, C15-SOA and C20-SOA account for less than 4% of the total SOA (Liu et al., 2024), this impact is also negligible.

We only implemented biogenic HOMs chemistry in the global model due to the limited knowledge of explicit chemical reactions forming anthropogenic-derived HOMs (Wang et al., 2017; Wang et al., 2020; Garmash et al., 2020; Molteni et al., 2018). This treatment likely leads to an underestimation of organic nucleation rates, particularly in urban areas (Fig. 1). More studies on chemical mechanisms of anthropogenic HOMs which could be applied in global model are needed. Our findings suggest that subsequent growth of the newly formed particles to larger sizes may have a more significant effect on aerosol number than nucleation. More studies are needed to quantify the contribution of anthropogenic organics to the initial growth rate. Changes in simulated aerosol number and size distribution caused by anthropogenic HOMs-driven NPF can have

important implications for CCN concentrations and aerosol indirect forcing (Wang and Penner, 2009; Wang et al., 2009; Gordon et al., 2016; Zhu et al., 2019), which also need further analysis.

Besides organic species, the concentration of inorganic nucleating species will also affect the accuracy of the total nucleation rate. The overestimation of H_2SO_4 in CAM6-Chem could potentially impact our final results regarding the organic propor-tion in both nucleation and the initial growth rate because both the dependencies of inorganic and organic nucleation rate on H_2SO_4 concentration are modeled with an exponent greater than 2 (Eq. (2)-(6)). Also, ammonia (NH_3) emissions used in this study are adapted from the Community Emissions Data System (CEDS), which remain challenging to represent in models due to uncertainties, particularly in specific sectors. NH_3 emissions from human waste were adapted using method-ologies from the Regional Emissions Inventory in Asia (REAS) (Kurokawa et al., 2013) and rely on a single global default emission factor. Not only is this emission factor uncertain, but there will certainly be regional variations due to differing environmental conditions that we were unable to take into account (Hoesly et al., 2018). For agricultural emissions, the ac-tual practices of managing livestock manure will affect true emissions; such practices vary significantly across the world but are not always well understood or reflected in the emission factors used in global inventories (Paulot et al., 2014). The aforementioned uncertainties in NH_3 will affect the inorganic nucleation rate and, consequently, the contribution of organics to the total nucleation rate.

In addition to the concentration of nucleating species, the uncertainty associated with NPF parameterization is also present in the model. Zhang et al. (2011) showed that radon contributes additional ionization in the boundary layer, especially over land. This implies that our pure organic nucleation rate might be underestimated since we only consider ion-pair production rate caused by galactic cosmic rays. This effect is negligible even over the continents since the contribution of ionization rate caused by the radioactive decay of radon is only significant (> 30%) within the lowest 1 km (Fig. 12 in Zhang et al. (2011)). Above 3 km, the contribution of radon decay induced ionization rate can be neglected (<10%). In this study, we focused on the proportion of organic NPF in the vertical integration within the whole atmosphere. Therefore, we will not consider incorporating the ion nucleation rate caused by radon.

The NPF rate at around 20 nm is calculated based on Eq. (14) from Kerminen and Kulmala (2002). This calculation is derived using several simplifying assumptions and approximations: (1) the only important sink for the newly formed particles is their coagulation with larger pre-existing particles; (2) the newly formed particles grow by condensation at a constant rate; (3) the pre-existing population of larger particles remains unchanged during the growth of the newly formed particles. However, Lehtinen et al. (2007) reformulated the previously published theory (Kerminen and Kulmala, 2002) to better account for the size dependence of the loss rates of newly formed particles (i.e., coagulation sink), rather than simplifying it as the gas condensation sink (CS). The uncertainty range caused by using the constant CS to replace the size-dependent coagulation sink (CoagS) is shown in Text S5. Recent studies (Stolzenburg et al., 2020; Ozon et al., 2021; Deng et al., 2020) also show that aerosol growth rates are not constant with size. However, CAM6-Chem

does not include a nucleation mode, which means that newly formed particles grow from 1.7 nm to 20 nm (geometric diameter in Aitken mode) within one physical timestep (30 minutes), making it impossible to resolve the growth rates of sub-20 nm particles. Future work, such as im-plementing a nucleation mode in CAM6-Chem and resolving particle growth rate within 20 nm, is therefore worth exploring."

Reference

Berndt, T., Mentler, B., Scholz, W., Fischer, L., Herrmann, H., Kulmala, M., and Hansel, A.: Accretion Product Formation from Ozonolysis and OH Radical Reaction of α-Pinene: Mechanistic Insight and the Influence of Isoprene and Ethylene, Environ. Sci. Technol., 52, 11069-11077, 10.1021/acs.est.8b02210, 2018.

Deng, C. J., Fu, Y. Y., Dada, L., Yan, C., Cai, R. L., Yang, D. S., Zhou, Y., Yin, R. J., Lu, Y. Q., Li, X. X., Qiao, X. H., Fan, X. L., Nie, W., Kontkanen, J., Kangasluoma, J., Chu, B. W., Ding, A. J., Kerminen, V. M., Paasonen, P., Worsnop, D. R., Bianchi, F., Liu, Y. C., Zheng, J., Wang, L., Kulmala, M., and Jiang, J. K.: Seasonal Characteristics of New Particle Formation and Growth in Urban Beijing, Environ. Sci. Technol., 54, 8547-8557, 10.1021/acs.est.0c00808, 2020.

Frege, C., Ortega, I. K., Rissanen, M. P., Praplan, A. P., Steiner, G., Heinritzi, M., Ahonen, L., Amorim, A., Bernhammer, A. K., Bianchi, F., Brilke, S., Breitenlechner, M., Dada, L., Dias, A., Duplissy, J., Ehrhart, S., El-Haddad, I., Fischer, L., Fuchs, C., Garmash, O., Gonin, M., Hansel, A., Hoyle, C. R., Jokinen, T., Junninen, H., Kirkby, J., Kürten, A., Lehtipalo, K., Leiminger, M., Mauldin, R. L., Molteni, U., Nichman, L., Petäjä, T., Sarnela, N., Schobesberger, S., Simon, M., Sipilä, M., Stolzenburg, D., Tomé, A., Vogel, A. L., Wagner, A. C., Wagner, R., Xiao, M., Yan, C., Ye, P., Curtius, J., Donahue, N. M., Flagan, R. C., Kulmala, M., Worsnop, D. R., Winkler, P. M., Dommen, J., and Baltensperger, U.: Influence of temperature on the molecular composition of ions and charged clusters during pure biogenic nucleation, Atmos. Chem. Phys., 18, 65-79, 10.5194/acp-18-65-2018, 2018.

Garmash, O., Rissanen, M. P., Pullinen, I., Schmitt, S., Kausiala, O., Tillmann, R., Zhao, D., Percival, C., Bannan, T. J., Priestley, M., Hallquist, Å. M., Kleist, E., Kiendler-Scharr, A., Hallquist, M., Berndt, T., McFiggans, G., Wildt, J., Mentel, T. F., and Ehn, M.: Multi-generation OH oxidation as a source for highly oxygenated organic molecules from aromatics, Atmos. Chem. Phys., 20, 515-537, 10.5194/acp-20-515-2020, 2020.

Gordon, H., Sengupta, K., Rap, A., Duplissy, J., Frege, C., Williamson, C., Heinritzi, M., Simon, M., Yan, C., Almeida, J., Trostl, J., Nieminen, T., Ortega, I. K., Wagner, R., Dunne, E. M., Adamov, A., Amorim, A., Bernhammer, A. K., Bianchi, F., Breitenlechner, M., Brilke, S., Chen, X. M., Craven, J. S., Dias, A., Ehrhart, S., Fischer, L., Flagan, R. C., Franchin, A., Fuchs, C., Guida, R., Hakala, J., Hoyle, C. R., Jokinen, T., Junninen, H., Kangasluoma, J., Kim, J., Kirkby, J., Krapf, M., Kurten, A., Laaksonen, A., Lehtipalo, K., Makhmutov, V., Mathot, S., Molteni, U., Monks, S. A., Onnela, A., Perakyla, O., Piel, F., Petaja, T., Praplanh, A. P., Pringle, K. J., Richards, N. A. D., Rissanen, M. P., Rondo, L., Sarnela, N., Schobesberger, S., Scott, C. E., Seinfeldo, J. H., Sharma, S., Sipila, M., Steiner, G., Stozhkov, Y., Stratmann, F., Tome, A., Virtanen, A., Vogel, A. L., Wagner, A. C., Wagner, P. E., Weingartner, E., Wimmer, D., Winkler, P. M., Ye, P. L., Zhang, X., Hansel, A., Dommen, J., Donahue, N. M., Worsnop, D. R., Baltensperger, U., Kulmala, M., Curtius, J., and Carslaw, K. S.: Reduced anthropogenic aerosol radiative forcing caused by biogenic new particle formation, P. Natl. Acad. Sci. USA, 113, 12053-12058, 10.1073/pnas.1602360113, 2016.

Heinritzi, M., Dada, L., Simon, M., Stolzenburg, D., Wagner, A. C., Fischer, L., Ahonen, L. R., Amanatidis, S., Baalbaki, R., Baccarini, A., Bauer, P. S., Baumgartner, B., Bianchi, F., Brilke, S., Chen, D., Chiu, R., Dias, A., Dommen, J., Duplissy, J., Finkenzeller, H., Frege, C., Fuchs, C., Garmash, O., Gordon, H., Granzin, M., El Haddad, I., He, X., Helm, J., Hofbauer, V., Hoyle, C. R., Kangasluoma, J., Keber, T., Kim, C., Kürten, A., Lamkaddam, H., Laurila, T. M., Lampilahti, J., Lee, C. P., Lehtipalo, K., Leiminger, M., Mai, H., Makhmutov, V., Manninen, H. E., Marten, R., Mathot, S., Mauldin, R. L., Mentler, B., Molteni, U., Müller, T., Nie, W., Nieminen, T., Onnela, A., Partoll, E., Passananti, M., Petäjä, T., Pfeifer, J., Pospisilova, V., Quéléver, L. L. J., Rissanen, M. P., Rose, C., Schobesberger, S., Scholz, W., Scholze, K., Sipilä, M., Steiner, G., Stozhkov, Y., Tauber, C., Tham, Y. J., Vazquez-Pufleau, M., Virtanen, A., Vogel, A. L., Volkamer, R., Wagner, R., Wang, M., Weitz, L., Wimmer, D., Xiao, M., Yan, C., Ye, P., Zha, Q., Zhou, X., Amorim, A., Baltensperger, U., Hansel, A., Kulmala, M., Tomé, A., Winkler, P. M., Worsnop, D. R., Donahue, N. M., Kirkby, J., and Curtius, J.: Molecular understanding of the suppression of new-particle formation by isoprene, Atmos. Chem. Phys., 20, 11809-11821, 10.5194/acp-20-11809-2020, 2020.

Hoesly, R. M., Smith, S. J., Feng, L., Klimont, Z., Janssens-Maenhout, G., Pitkanen, T., Seibert, J. J., Vu, L., Andres, R. J., Bolt, R. M., Bond, T. C., Dawidowski, L., Kholod, N., Kurokawa, J. I., Li, M., Liu, L., Lu, Z., Moura, M. C. P., O'Rourke, P. R., and Zhang, Q.: Historical (1750–2014) anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data System (CEDS), Geosci. Model Dev., 11, 369-408, 10.5194/gmd-11-369-2018, 2018.

Kerminen, V. M. and Kulmala, M.: Analytical formulae connecting the "real" and the "apparent" nucleation rate and the nuclei number concentration for atmospheric nucleation events, J. Atmos. Sci., 33, 609-622, 10.1016/s0021-8502(01)00194-x, 2002.

Kurokawa, J., Ohara, T., Morikawa, T., Hanayama, S., Janssens-Maenhout, G., Fukui, T., Kawashima, K., and Akimoto, H.: Emissions of air pollutants and greenhouse gases over Asian regions during 2000–2008: Regional Emission inventory in ASia (REAS) version 2, Atmos. Chem. Phys., 13, 11019-11058, 10.5194/acp-13-11019-2013, 2013.

Lehtinen, K. E. J., Dal Maso, M., Kulmala, M., and Kerminen, V.-M.: Estimating nucleation rates from apparent particle formation rates and vice versa: Revised formulation of the Kerminen–Kulmala equation, J. Aerosol Sci., 38, 988-994, https://doi.org/10.1016/j.jaerosci.2007.06.009, 2007.

Liu, Y., Dong, X., Wang, M., Xu, R., Thornton, J. A., Shao, X., Emmons, L. K., Jo, D. S., Yue, M., and Shrivastava, M.: A Modeling Study of Global Distribution and Formation Pathways of Highly Oxygenated Organic Molecules Derived Secondary Organic Aerosols (HOMs-SOA) from Monoterpenes, (under review), 2024.

Molteni, U., Bianchi, F., Klein, F., El Haddad, I., Frege, C., Rossi, M. J., Dommen, J., and Baltensperger, U.: Formation of highly oxygenated organic molecules from aromatic compounds, Atmos. Chem. Phys., 18, 1909-1921, 10.5194/acp-18-1909-2018, 2018.

Ozon, M., Stolzenburg, D., Dada, L., Seppänen, A., and Lehtinen, K. E. J.: Aerosol formation and growth rates from chamber experiments using Kalman smoothing, Atmos. Chem. Phys., 21, 12595-12611, 10.5194/acp-21-12595-2021, 2021.

Paulot, F., Jacob, D. J., Pinder, R. W., Bash, J. O., Travis, K., and Henze, D. K.: Ammonia emissions in the United States, European Union, and China derived by high-resolution inversion of ammonium wet deposition data: Interpretation with a new agricultural emissions inventory (MASAGE_NH3), J. Geophys. Res.-Atmos., 119, 4343-4364, https://doi.org/10.1002/2013JD021130, 2014.

Rissanen, M. P., Kurtén, T., Sipilä, M., Thornton, J. A., Kausiala, O., Garmash, O., Kjaergaard, H. G., Petäjä, T., Worsnop, D. R., Ehn, M., and Kulmala, M.: Effects of Chemical Complexity on the Autoxidation Mechanisms of Endocyclic Alkene Ozonolysis Products: From Methylcyclohexenes toward Understanding α-Pinene, J. Phys. Chem. A, 119, 4633-4650, 10.1021/jp510966g, 2015.

Schervish, M. and Donahue, N. M.: Peroxy radical chemistry and the volatility basis set, Atmospheric Chemistry and Physics, 20, 1183-1199, 10.5194/acp-20-1183-2020, 2020.

Simon, M., Dada, L., Heinritzi, M., Scholz, W., Stolzenburg, D., Fischer, L., Wagner, A. C., Kürten, A., Rörup, B., He, X. C., Almeida, J., Baalbaki, R., Baccarini, A., Bauer, P. S., Beck, L., Bergen, A., Bianchi, F., Bräkling, S., Brilke, S., Caudillo, L., Chen, D., Chu, B., Dias, A., Draper, D. C., Duplissy, J., El-Haddad, I., Finkenzeller, H., Frege, C., Gonzalez-Carracedo, L., Gordon, H., Granzin, M., Hakala, J., Hofbauer, V., Hoyle, C. R., Kim, C., Kong, W., Lamkaddam, H., Lee, C. P., Lehtipalo, K., Leiminger, M., Mai, H., Manninen, H. E., Marie, G., Marten, R., Mentler, B., Molteni, U., Nichman, L., Nie, W., Ojdanic, A., Onnela, A., Partoll, E., Petäjä, T., Pfeifer, J., Philippov, M., Quéléver, L. L. J., Ranjithkumar, A., Rissanen, M. P., Schallhart, S., Schobesberger, S., Schuchmann, S., Shen, J., Sipilä, M., Steiner, G., Stozhkov, Y., Tauber, C., Tham, Y. J., Tomé, A. R., Vazquez-Pufleau, M., Vogel, A. L., Wagner, R., Wang, M., Wang, D. S., Wang, Y., Weber, S. K., Wu, Y., Xiao, M., Yan, C., Ye, P., Ye, Q., Zauner-Wieczorek, M., Zhou, X., Baltensperger, U., Dommen, J., Flagan, R. C., Hansel, A., Kulmala, M., Volkamer, R., Winkler, P. M., Worsnop, D. R., Donahue, N. M., Kirkby, J., and Curtius, J.: Molecular understanding of new-particle formation from α-pinene between −50 and +25 °C, Atmos. Chem. Phys., 20, 9183-9207, 10.5194/acp-20-9183-2020, 2020.

Stolzenburg, D., Fischer, L., Vogel, A. L., Heinritzi, M., Schervish, M., Simon, M., Wagner, A. C., Dada, L., Ahonen, L. R., Amorim, A., Baccarini, A., Bauer, P. S., Baumgartner, B., Bergen, A., Bianchi, F., Breitenlechner, M., Brilke, S., Buenrostro Mazon, S., Chen, D., Dias, A., Draper, D. C., Duplissy, J., El Haddad, I., Finkenzeller, H., Frege, C., Fuchs, C., Garmash, O., Gordon, H., He, X., Helm, J., Hofbauer, V., Hoyle, C. R., Kim, C., Kirkby, J., Kontkanen, J., Kürten, A., Lampilahti, J., Lawler, M., Lehtipalo, K., Leiminger, M., Mai, H., Mathot, S., Mentler, B., Molteni, U., Nie, W., Nieminen, T., Nowak, J. B., Ojdanic, A., Onnela, A., Passananti, M., Petäjä, T., Quéléver, L. L. J., Rissanen, M. P., Sarnela, N., Schallhart, S., Tauber, C., Tomé, A., Wagner, R., Wang, M., Weitz, L., Wimmer, D., Xiao, M., Yan, C., Ye, P., Zha, Q., Baltensperger, U., Curtius, J., Dommen, J., Flagan, R. C., Kulmala, M., Smith, J. N., Worsnop, D. R., Hansel, A., Donahue, N. M., and Winkler, P. M.: Rapid growth of organic aerosol nanoparticles over a wide tropospheric temperature range, P. Natl. Acad. Sci. USA, 115, 9122-9127, 10.1073/pnas.1807604115, 2018.

Stolzenburg, D., Simon, M., Ranjithkumar, A., Kurten, A., Lehtipalo, K., Gordon, H., Ehrhart, S., Finkenzeller, H., Pichelstorfer, L., Nieminen, T., He, X. C., Brilke, S., Xiao, M., Amorim, A., Baalbaki, R., Baccarini, A., Beck, L., Brakling, S., Murillo, L. C., Chen, D. X., Chu, B. W., Dada, L., Dias, A., Dommen, J., Duplissy, J., El Haddad, I., Fischer, L., Carracedo, L. G., Heinritzi, M., Kim, C., Koenig, T. K., Kong, W., Lamkaddam, H., Lee, C. P., Leiminger, M., Li, Z. J., Makhmutov, V., Manninen, H. E., Marie, G., Marten, R., Muller, T., Nie, W., Partoll, E., Petaja, T., Pfeifer, J., Philippov, M., Rissanen, M. P., Rorup, B., Schobesberger, S., Schuchmann, S., Shen, J. L., Sipila, M., Steiner, G.,

Stozhkov, Y., Tauber, C., Tham, Y. J., Tome, A., Vazquez-Pufleau, M., Wagner, A. C., Wang, M. Y., Wang, Y. H., Weber, S. K., Wimmer, D., Wlasits, P. J., Wu, Y. S., Ye, Q., Zauner-Wieczorek, M., Baltensperger, U., Carslaw, K. S., Curtius, J., Donahue, N. M., Flagan, R. C., Hansel, A., Kulmala, M., Lelieveld, J., Volkamer, R., Kirkby, J., and Winkler, P. M.: Enhanced growth rate of atmospheric particles from sulfuric acid, Atmos. Chem. Phys., 20, 7359-7372, 10.5194/acp-20-7359-2020, 2020.

Wang, M. and Penner, J. E.: Aerosol indirect forcing in a global model with particle nucleation, Atmos. Chem. Phys., 9, 239-260, 10.5194/acp-9-239-2009, 2009.

Wang, M., Penner, J. E., and Liu, X.: Coupled IMPACT aerosol and NCAR CAM3 model: Evaluation of predicted aerosol number and size distribution, J. Geophys. Res.-Atmos.,, 114, 10.1029/2008jd010459, 2009.

Wang, S. N., Wu, R. R., Berndt, T., Ehn, M., and Wang, L. M.: Formation of Highly Oxidized Radicals and Multifunctional Products from the Atmospheric Oxidation of Alkylbenzenes, Environ. Sci. Technol., 51, 8442-8449, 10.1021/acs.est.7b02374, 2017.

Wang, Y., Mehra, A., Krechmer, J. E., Yang, G., Hu, X., Lu, Y., Lambe, A., Canagaratna, M., Chen, J., Worsnop, D., Coe, H., and Wang, L.: Oxygenated products formed from OH-initiated reactions of trimethylbenzene: autoxidation and accretion, Atmos. Chem. Phys., 20, 9563-9579, 10.5194/acp-20-9563-2020, 2020.

Ye, Q., Wang, M., Hofbauer, V., Stolzenburg, D., Chen, D., Schervish, M., Vogel, A., Mauldin, R. L., Baalbaki, R., Brilke, S., Dada, L., Dias, A., Duplissy, J., El Haddad, I., Finkenzeller, H., Fischer, L., He, X., Kim, C., Kürten, A., Lamkaddam, H., Lee, C. P., Lehtipalo, K., Leiminger, M., Manninen, H. E., Marten, R., Mentler, B., Partoll, E., Petäjä, T., Rissanen, M., Schobesberger, S., Schuchmann, S., Simon, M., Tham, Y. J., Vazquez-Pufleau, M., Wagner, A. C., Wang, Y., Wu, Y., Xiao, M., Baltensperger, U., Curtius, J., Flagan, R., Kirkby, J., Kulmala, M., Volkamer, R., Winkler, P. M., Worsnop, D., and Donahue, N. M.: Molecular Composition and Volatility of Nucleated Particles from α-Pinene Oxidation between −50 °C and +25 °C, Environ. Sci. Technol., 53, 12357-12365, 10.1021/acs.est.9b03265, 2019.

Zhang, Y. M., Zhang, X. Y., Sun, J. Y., Lin, W. L., Gong, S. L., Shen, X. J., and Yang, S.: Characterization of new particle and secondary aerosol formation during summertime in Beijing, China, Tellus B., 63, 382-394, 10.1111/j.1600-0889.2011.00533.x, 2011.

Zhao, D., Schmitt, S. H., Wang, M., Acir, I. H., Tillmann, R., Tan, Z., Novelli, A., Fuchs, H., Pullinen, I., Wegener, R., Rohrer, F., Wildt, J., Kiendler-Scharr, A., Wahner, A., and Mentel, T. F.: Effects of NOx and SO2 on the secondary organic aerosol formation from photooxidation of α-pinene and limonene, Atmos. Chem. Phys., 18, 1611-1628, 10.5194/acp-18-1611-2018, 2018.

Zhu, J. L., Penner, J. E., Yu, F. Q., Sillman, S., Andreae, M. O., and Coe, H.: Decrease in radiative forcing by organic aerosol nucleation, climate, and land use change, Nat.Commun., 10, 10.1038/s41467-019-08407-7, 2019.

I noticed two technical issues in this section: first, nitrate is a secondary species and CMIP6 inventories provide NOx which reacts and partitions to aerosol to make nitrate. So it is not meaningful to comment that CMIP6 inventories do not include nitrate.

Response: We are grateful to the referee for pointing out this technical issue. We have revised the manuscript to ensure that the discussion accurately reflects uncertainties in the concentrations of NH_3. The following discussion have been included in the **Summary**:

"Ammonia (NH_3) emissions used in this study are adapted from the Community Emissions Data System (CEDS), which remain challenging to represent in models due to uncertainties, particularly in specific sectors. NH_3 emissions from human waste were adapted using methodologies from the Regional Emissions Inventory in Asia (REAS) (Kurokawa et al., 2013) and rely on a single global default emission factor. Not only is this emission factor uncertain, but there will certainly be regional variations due to differing environmental conditions that we were unable to take into account (Hoesly et al., 2018). For agricultural emissions, the actual practices of managing livestock manures will affect true emissions; such practices vary significantly across the world but are not always well understood or reflected in the emission factors used in global inventories (Paulot et al., 2014). The aforementioned uncertainties in NH_3 will affect the inorganic nucleation rate and, consequently, the contribution of organics to the total nucleation rate."

Reference

Kurokawa, J., Ohara, T., Morikawa, T., Hanayama, S., Janssens-Maenhout, G., Fukui, T., Kawashima, K., and Akimoto, H.: Emissions of air pollutants and greenhouse gases over Asian regions during 2000–2008: Regional Emission inventory in ASia (REAS) version 2, Atmos. Chem. Phys., 13, 11019-11058, 10.5194/acp-13-11019-2013, 2013.

Hoesly, R. M., Smith, S. J., Feng, L., Klimont, Z., Janssens-Maenhout, G., Pitkanen, T., Seibert, J. J., Vu, L., Andres, R. J., Bolt, R. M., Bond, T. C., Dawidowski, L., Kholod, N., Kurokawa, J. I., Li, M., Liu, L., Lu, Z., Moura, M. C. P., O'Rourke, P. R., and Zhang, Q.: Historical (1750–2014) anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data System (CEDS), Geosci. Model Dev., 11, 369-408, 10.5194/gmd-11-369-2018, 2018.

Paulot, F., Jacob, D. J., Pinder, R. W., Bash, J. O., Travis, K., and Henze, D. K.: Ammonia emissions in the United States, European Union, and China derived by high-resolution inversion of ammonium wet deposition data: Interpretation with a new agricultural emissions inventory (MASAGE_NH3), J. Geophys. Res.-Atmos., 119, 4343-4364, https://doi.org/10.1002/2013JD021130, 2014.

Second, it is not clear to me that it is impossible to implement the equation of Lehtinen et al (2007) in the absence of a nucleation mode? The coagulation sink can still be approximated whether or not such a mode exists.

Response: We apologize for any misunderstanding caused by not clearly explaining the details of the issue. Our intended meaning is that we cannot calculate the aerosol loss rate at any diameter between 1.7 nm and 20 nm using Equation (5) in Lehtinen et al. (2007) because there is no calculation of the aerosol coagulation sink at 1 nm in the Aitken mode of CAM6-Chem.

However, we recognize that these potentially large uncertainties are important and should be addressed through sensitivity studies. Without altering the model structure, we tested the uncertainty range in the aerosol loss rate calculation caused by using a constant gas condensation sink (CS) instead of the size-dependent coagulation sink (CoagS) by applying Equation (8) from Lehtinen et al. (2007) in **Test S5** and have also added the corresponding discussion in the **Summary**.

Text S5. Sensitivity Test of the Loss Rate of Newly Formed Particles

"In order to evaluate the uncertainty range caused by using a constant gas condensation sink (CS) instead of a size-dependent coagulation sink (CoagS) to calculate apparent nucleation rate ($j_{20 \text{ nm}}$), Eq. (8) in Lehtinen et al. (2007) was modified as follows to calculate the ratio of CS to CoagS at 1.7 nm:

$$\frac{CS}{CoagS\ (1.7\text{nm})} = (\frac{0.71}{1.7})^{-m} \qquad (S6)$$

The exponent m has a value, depending on the background distribution, between −1 and −2. For typical atmospheric aerosols, the value of m is expected to be between −1.5 and −1.9 (Lehtinen et al., 2007) and results of the CS/CoagS (1.7 nm) ratio using different m values are shown in **Table S3**."

Table S3. The ratio of the gas condensation sink (CS) to the aerosol coagulation sink at 1.7 nm (CoagS (1.7 nm)) during NPF events for different m values provided by Lehtinen et al. (2007).

m values	CS / CoagS (1.7nm)
1.5	3.63
1.6	3.98
1.7	4.40
1.8	4.91
1.9	5.51

We also added the following discussion (**<u>Underlined</u>**) at the end of the original paragraph (in **Blue**, without underlining) in the **Summary**:

"The NPF rate at around 20 nm is calculated based on Eq. (14) based on Kerminen and Kulmala (2002). It is derived with several simplifying assumptions and approximations: (1) the only important sink for the newly formed particles is their coagulation with larger pre-existing particles; (2) the newly formed particles grow by condensation at a constant rate; (3) the pre-existing population of larger particles remains unchanged during the growth of the newly formed particles. However, Lehtinen et al. (2007) reformulated the previously published theory (Kerminen and Kulmala, 2002) to better account for the size dependence of the loss rates

of newly formed particles (i.e., coagulation sink), rather than simplifying it as the gas condensation sink (CS). The error analysis of the size-dependent coagulation sink (CoagS) compared to the constant CoagS (simplified as gas CS) is shown in Text S5. Recent studies (Stolzenburg et al, 2020; Ozon et al. 2021; Deng et al., 2020) also show that aerosol growth rates are not constant with size. However, CAM6-Chem does not include a nucleation mode, which means that newly formed particles grow from 1.7 nm to 20 nm (geometric diameter in Aitken mode) within one physical timestep (30 minutes), making it impossible to resolve the growth rates of sub-20 nm particles. Future work, such as implementing a nucleation mode in CAM6-Chem and resolving particle growth rates within 20 nm, is worth exploring."

Reference

Kerminen, V. M. and Kulmala, M.: Analytical formulae connecting the "real" and the "apparent" nucleation rate and the nuclei number concentration for atmospheric nucleation events, J. Atmos. Sci., 33, 609-622, 10.1016/s0021-8502(01)00194-x, 2002.

Lehtinen, K. E. J., Dal Maso, M., Kulmala, M., and Kerminen, V.-M.: Estimating nucleation rates from apparent particle formation rates and vice versa: Revised formulation of the Kerminen–Kulmala equation, J. Aerosol Sci., 38, 988-994, https://doi.org/10.1016/j.jaerosci.2007.06.009, 2007.

Stolzenburg, D., Simon, M., Ranjithkumar, A., Kurten, A., Lehtipalo, K., Gordon, H., Ehrhart, S., Finkenzeller, H., Pichelstorfer, L., Nieminen, T., He, X. C., Brilke, S., Xiao, M., Amorim, A., Baalbaki, R., Baccarini, A., Beck, L., Brakling, S., Murillo, L. C., Chen, D. X., Chu, B. W., Dada, L., Dias, A., Dommen, J., Duplissy, J., El Haddad, I., Fischer, L., Carracedo, L. G., Heinritzi, M., Kim, C., Koenig, T. K., Kong, W., Lamkaddam, H., Lee, C. P., Leiminger, M., Li, Z. J., Makhmutov, V., Manninen, H. E., Marie, G., Marten, R., Muller, T., Nie, W., Partoll, E., Petaja, T., Pfeifer, J., Philippov, M., Rissanen, M. P., Rorup, B., Schobesberger, S., Schuchmann, S., Shen, J. L., Sipila, M., Steiner, G., Stozhkov, Y., Tauber, C., Tham, Y. J., Tome, A., Vazquez-Pufleau, M., Wagner, A. C., Wang, M. Y., Wang, Y. H., Weber, S. K., Wimmer, D., Wlasits, P. J., Wu, Y. S., Ye, Q., Zauner-Wieczorek, M., Baltensperger, U., Carslaw, K. S., Curtius, J., Donahue, N. M., Flagan, R. C., Hansel, A., Kulmala, M., Lelieveld, J., Volkamer, R., Kirkby, J., and Winkler, P. M.: Enhanced growth rate of atmospheric particles from sulfuric acid, Atmos. Chem. Phys., 20, 7359-7372, 10.5194/acp-20-7359-2020, 2020.

Ozon, M., Stolzenburg, D., Dada, L., Seppänen, A., and Lehtinen, K. E. J.: Aerosol formation and growth rates from chamber experiments using Kalman smoothing, Atmos. Chem. Phys., 21, 12595-12611, 10.5194/acp-21-12595-2021, 2021.

Deng, C. J., Fu, Y. Y., Dada, L., Yan, C., Cai, R. L., Yang, D. S., Zhou, Y., Yin, R. J., Lu, Y. Q., Li, X. X., Qiao, X. H., Fan, X. L., Nie, W., Kontkanen, J., Kangasluoma, J., Chu, B. W., Ding, A. J., Kerminen, V. M., Paasonen, P., Worsnop, D. R., Bianchi, F., Liu, Y. C., Zheng, J., Wang, L., Kulmala, M., and Jiang, J. K.: Seasonal Characteristics of New Particle Formation and Growth in Urban Beijing, Environ. Sci. Technol., 54, 8547-8557, 10.1021/acs.est.0c00808, 2020.