
Response to the Anonymous Referee of the manuscript “Iodine oxoacids and their roles in sub-3 nanometer particle growth in polluted urban environments”

Response to Anonymous Referee #1:

This study reports unique long-term CIMS measurements of HIO_x as well as NPF events in two urban areas, Beijing and Nanjing, China. The source analysis of iodic acid indicated potential long-range transport from marine and terrestrial sources rather than local anthropogenic emissions. They also estimated the contribution of HIO_x and H₂SO₄ to nanoparticle growth rate and survival probability of nanoparticles. This paper shows that, although iodic acid does not play a major role in nanoparticle growth rate, the additional growth by iodic acid can enhance the survival of sub-3 nm particles in Beijing during NPF events. This suggests iodic acid contributes to NPF events in urban conditions with support from field measurements. However, some technical details on the measurement and analysis need to be clarified. Therefore, I recommend the manuscript for publication after revisions to address my comments below.

We thank the Reviewer for carefully checking our manuscript and giving us helpful and constructive comments pointing us towards some important improvements in our analysis. Below we give a point-by-point response including the changes we made to the revised version of the manuscript. Comments are shown as *blue italic text* followed by our responses. Changes made to the revised manuscript are **highlighted** and shown as “quoted underlined text” in our responses.

General revision:

We have renumbered all the supplementary figures according to their orders of appearance in the main text.

(1) Eq. (9) & (11): What was the range of temperature/size correction factor applied to GR(HIO₃) in eq. (9)? Also, please explain why GR(H₂SO₄) in eq. (11) does not need temperature correction. In eq. (5), GR is corrected only to account for growth by condensation. And it is unclear in the text whether GR(HIO₃) and GR(H₂SO₄) in those equations are growth rates only by condensation (excluding coagulation).

Response: Thanks for the comment. For GR(HIO₃) calculation, which denotes the growth rates contributed by iodic acid (HIO₃), we adopted the CLOUD chamber experiment results obtained by

He et al. (2021). The growth rate measurements were carried out at 10 °C for the growth of 1.8-3.2 nm particles. Therefore, we corrected the fitted GR for both the temperature and size range difference using the Eq. (10). As depicted in Fig. R1, the correction factor for GR(HIO₃) is rather small (close to one) to both size range selection and temperature variation. The correction factor applied to field measurements at two sites and CLOUD chamber are close to 1. We further tested a larger temperature range from 10 to 50 °C in Fig. R1(b) and investigated the correction factors for the two size ranges studied here. The correction factors stay robust which suggests that the temperature and size range differences between the CLOUD experiments and field observations remain minimal. To further elaborate the calculations, the manuscript has been revised as below,

[Line 256-257] “To better understand the role of HIO₃ as an additional GR contributor at two sites, we calculate the condensational GR of HIO₃ and H₂SO₄ during NPF events, the computation criteria are listed as below.”

[Line 277-281] “The correction factors remained consistently close to 1 (as shown in Fig. S2 for more detailed information). This suggests that the variations in size range and temperature between the CLOUD measurements and our field observations are minimal.”

[Line 292-294] “H₂SO₄ contribution to GR is calculated as a first-order approximation independent of temperature as Eq. (11) (Stolzenburg et al., 2020), where $d_p = \frac{d_{p_{initial}} + d_{p_{final}}}{2}$ in nm, and the subscripts initial and final refer to the particle diameter at the beginning and the end of the growing process.”

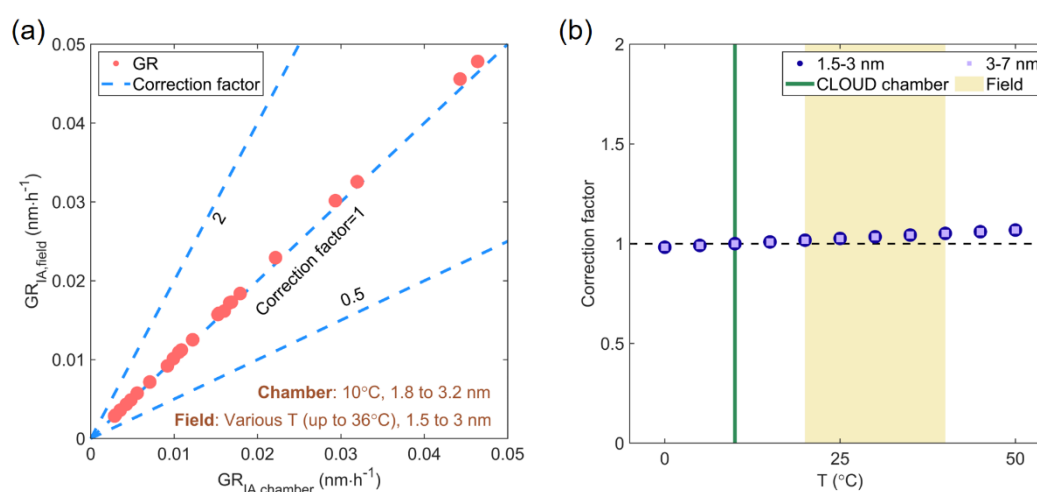


Figure R1. The calculated correction factors based on Eq. (10) and the sensitivity against temperature and size. (a) The correction factors derived based on chamber and field site measurement conditions during NPF events. (b) The correction factor as a function of temperature with blue circles and grey squares denoting 1.5-3 nm and 3-7 nm growth, respectively. Vertical green line indicates the temperature of the CLOUD experiments (10 °C) and the yellow shadow indicates the temperature range of field observations.

(2) Line 273-274: “We define SP_{tot} as the particle survival probability calculated using the measured GRs (in Beijing) or the expected growth rate considering growth contributions from both H_2SO_4 and HIO_3 (in Nanjing).” Please explain how using different inputs for SP_{tot} calculation would affect EF in each site.

Response: Thanks for the comment. We adopt different procedures here to derive the enhancement factor (EF) of survival probability (SP) because the growth trajectory of sub-3 nm particles is difficult to capture at SORPES due to the limitation of instruments. Furthermore, considering that H_2SO_4 is the dominant vapor for sub-3 nm range growth in urban environments (Deng et al., 2020a), we then let SP_{tot} at SORPES equal to the SP considering both contributions from H_2SO_4 and HIO_3 . Therefore, Eq. (12) for SORPES scenarios can be clarified as

$$EF = \frac{SP(GR_{H_2SO_4} + GR_{HIO_3})}{SP(GR_{H_2SO_4})} - 1. \quad (R1)$$

As shown in Fig. S13, the amplification of SP is especially sensitive against initial GR ($GR_{H_2SO_4}$ in this case). The $GR_{H_2SO_4}$ and GR_{HIO_3} at SORPES is derived based on the empirical equation and therefore, the measurement of two acids will introduce uncertainties for EF calculation at SORPES. On the contrary, the biggest uncertainty for EF calculation at BUCT/AHL will be the fitting process for measured GR in NPF events for sub-3 nm particle growth. Besides, the EF for 3-7 nm range growth is also calculated at BUCT/AHL but not at SORPES.

The values of EF for sub-3 nm particle growth at both sites can be checked via Table S7 and Table S9.

(3) Line 305-306: “The calibrated HIO_x concentration is above the detection limit during almost the entire measurement periods...” However, I can not find more details on the calibration, except for the general description of nitrate CIMS in section 2.1.2. Therefore, the quantification method of HIO_x (as well as H_2SO_4) needs to be shown and the sources of uncertainty need to be discussed since HIO_x concentrations are being used for the growth rate and survival probability calculations. How and how often were the instruments calibrated for HIO_x ? Do authors expect losses in the sampling line or variations of HIO_x sensitivity due to changes in temperature/relative humidity?

Response: Thanks for the suggestions. We have now included a brief discussion of the calibration processes, the references and the expected systematic error of our calibration method. And the manuscript has been revised as below,

[Line 135-146] “ H_2SO_4 calibration was conducted using a standardized method (Kurten et al., 2012; He et al., 2023). In a nutshell, the calibration of H_2SO_4 involved the reaction of an excessive amount of sulfur dioxide (SO_2) with a known quantity of hydroxyl (OH) radicals generated by a portable

mercury lamp. This mercury lamp is equipped with a filter to intercept the sample air containing water, which, in turn, is photolyzed to produce OH radicals. The convection-diffusion-reaction processes within the chemical ionization inlet can be accurately simulated using a two-dimensional model (e.g., the MARFORCE-Flowtube model) (He et al., 2023), allowing for the quantification of H₂SO₄ concentration at the mass spectrometer's entrance. The quantification of the measured signals for H₂SO₄ monomer at both sites are seasonally calibrated with diffusion losses in the inlet tube taken into consideration. Since both H₂SO₄ and HIO_x are detected at the collision limit, they share the same calibration factor (He et al., 2021; He et al., 2023). The general systematic error for the detection of H₂SO₄ and HIO_x is expected to be within 50% to 200% for field observations (Liu et al., 2021).”

(4) Line 426: “implies that marine iodine sources could be important” Could it be emissions from industrial areas along with the back trajectory path?

Response: Thanks for pointing out potential iodine sources. We use the back-trajectory analysis in this study to investigate the potential sources of HIO₃ precursor because of the limited knowledge of their sources in inland cities. Marine environments emit both inorganic iodine through ozone-stimulated liberation of HOI and I₂ from surface ocean and organic iodine compounds through biotic and abiotic processes such as CH₃I and CH₂I₂. In general, the inorganic iodine molecules, HOI and I₂, dominate the global iodine sources (Carpenter et al., 2013; Ordóñez et al., 2012). The measurement site of BUCT/AHL is 150 km away from the nearest coastline on the southeast side, therefore, we link the trajectory cluster from the same direction with marine sources, which accounts for the majority of level 1 clusters.

As for the industrial sources of iodine, a previous work conducted in urban Beijing found that intensive coal combustions are sources of atmospheric iodine during heating seasons (Shi et al., 2021), in the form of organic iodine compounds in northern China. However, the cluster analysis for back-trajectories at BUCT/AHL are specially for warm months (from May to September), so it can be inferred that the industrial emission of iodine compounds is less significant compared to the dominant iodine source of oceans.

We have specified the time window for back-trajectory analysis in Beijing, and the modified manuscript is shown as below,

[Line 458-460] “In order to investigate the source of HIO_x in urban environments, we further conduct cluster analysis of the air mass backward trajectories of the AHL/BUCT station in warm seasons (from May to September in this study).”

(5) Line 428: That is the opposite direction of continental outflow.

Response: Thank you for your comment. The northern cluster instead of the southern one would indicate the continental outflow better. Therefore, we corrected the expression in manuscript as following, according to your reminder.

[Line 465-466] “Additionally, the air mass travels from northern wind may also carry substantial precursors of HIO₃, indicating the potential terrestrial sources of iodine.”

(6) Line 445: Even if the HIO₃ emission (or secondary formation) is higher in winter (when PM_{2.5} loading is higher), still the concentration of HIO₃ can be lower due to potentially higher CS. Thus the y-axis in Fig.3 can be normalized by CS to see if the source of HIO₃ is correlated to PM_{2.5} pollution.

Response: Thank you for the good suggestions. Taken the steady-state assumption of ambient HIO₃, we get to evaluate the production of HIO₃ according to its loss rate, which is dominantly condensational loss. Therefore, the product of HIO₃ and CS is used to describe the HIO₃ production rate here and its correlation with PM_{2.5} is shown as Fig. R2. The results in the following figure show that there is no apparent impact of PM_{2.5} level on HIO₃ production. To avoid the seasonal variation of PM_{2.5}, we particularly investigate the correlation between HIO₃ production and PM_{2.5} in warm seasons in Fig. R2(b), and there is no significant correlation, either. Additionally, as depicted in Fig. R3, the relationship of CS and PM_{2.5} is further examined, which shows a significant positive relationship. Therefore, if PM_{2.5} pollution does contribute to the production of HIO₃, we would have expected to see the accordingly higher values of HIO₃*CS, because of higher CS and assumingly higher HIO₃ concentration. However, as depicted in Fig. R2, under high level of PM_{2.5} pollution, there is no significant elevation of HIO₃ production. To conclude, the production of HIO₃ is not correlated to PM_{2.5} pollution based on our measurements.

The relationship between HIO₃ production rate and PM_{2.5} have been mentioned in the revised manuscript as following,

[Line 486-488] “Furthermore, the results in Fig. S7 demonstrate that PM_{2.5} pollution does not play a conclusive role on the HIO₃ production, especially with the seasonal influence excluded.”

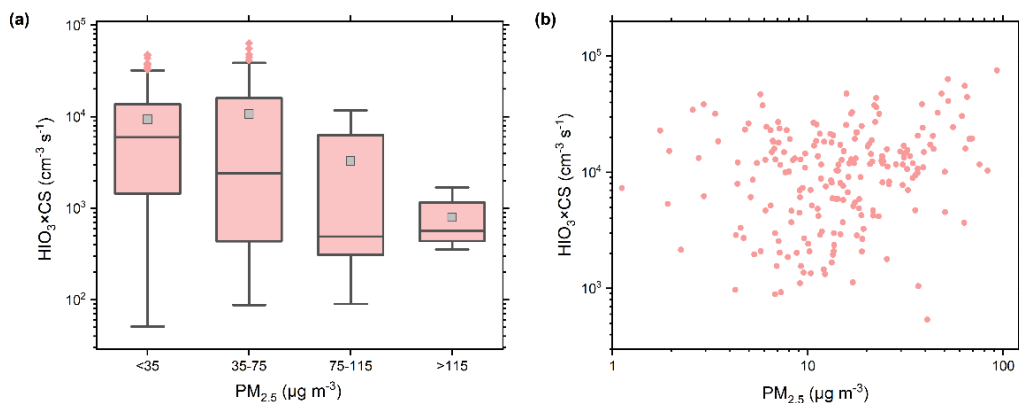


Figure R2. The impact of $\text{PM}_{2.5}$ on HIO_3 production calculated from the HIO_3 concentration and CS (a) in all seasons and (b) specifically in warm seasons (from May to September).

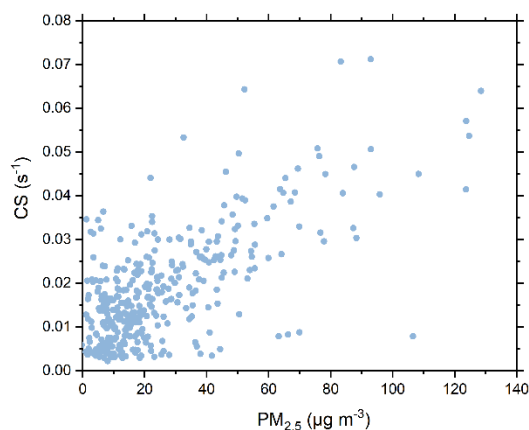


Figure R3. The correlation between CS and $\text{PM}_{2.5}$.

(7) Line 522-529: Survival parameter (P) is newly introduced here and it is unclear why P is being used for AHL/BUCT case while survival probability (SP) is used for the rest of the analysis. Please compare the survival parameter from Kulmala et al. (2017) and survival probability (SP) from Lehtinen et al. (2007) and explain why using P is useful here.

Response: Thanks for your good comments. The primary role of the survival parameter P (Kulmala et al., 2017) was to provide a viewpoint for the occurrence of NPF, similar to L or L_T (McMurry et al., 2005; Kuang et al., 2010; Cai et al., 2017), especially in urban environments. Conversely, survival probability (SP) serves as a quantitative estimate of what is the percentage of the recently formed particles that grows to larger sizes.

The survival parameter P was used here to characterize the particle formation events. Events with P smaller than 100 were selected in this study as the particle formation and growth under low CS conditions are more certain (Kulmala et al., 2017). When the P is larger than 100, the mechanisms of particle formation and growth under high CS conditions is still under debate (Kulmala et al., 2017) and thus these data are avoided.

Given that measured GRs are not derived due to the absence of sub-3 nm particle measurement, the survival parameter P is not calculated for SORPES station.

(8) Line 567: “a fixed GR enhancement” What does “fixed” mean?

Response: Our apologies for not being clear. In this section, we were trying to discuss about the sensitivity of SP enhancement against GR enhancement, which is controlled by CoagS.

For Eq. (8), by considering that d_{p1} , d_{p2} and CoagS are constant, we can further combine the constant values besides CoagS as a new term A .

Where

$$A = \frac{d_{p1}}{m-1} \left[\left(\frac{d_{p2}}{d_{p1}} \right)^{1-m} - 1 \right], \quad (R2)$$

and Eq. (8) can be simplified as

$$SP = \exp\left(\frac{CoagS}{A} \cdot \frac{1}{GR}\right). \quad (R3)$$

By assuming $m=1.7$, it can be deduced that $A < 0$, and if we take the derivative of SP with respect to GR for Eq. (R3), we further get the following Eq. (R4):

$$\frac{\Delta SP}{\Delta GR} = -\frac{1}{GR^2} \cdot \frac{CoagS}{A} \cdot SP. \quad (R4)$$

For a given GR value and GR enhancement (ΔGR), the enhancement of SP (ΔSP) is subject to the value of CoagS. The larger the CoagS is, the larger enhancement of SP can be expected.

For the two cases studied in this section, the GR values are similar while on Jun 21, 2021, the CoagS is 5 times larger than May 29, 2021, leading to a more pronounced enhancement of SP on the former day. The expressions we were using was somehow confusing, and we have revised the statement as below,

[Line 608-609] “This suggests that in polluted environments with higher CoagS, such as Beijing,

SP enhancement can be more sensitive to GR enhancement.”

(9) Line 570-575: Are the MOD, APT-x, and APT-y methods equally credible? Then the differences in those methods represent the uncertainty of using MOD fitting method? Or do APT-x and APT-y represent upper/lower limit of MOD fitting?

Response: Thank you for your comments. We acknowledge the need for clearer articulation regarding the methods used for GR calculations. To provide a more comprehensive explanation, we have expanded upon these methods in the revised supplemental information in [Line 1296-1317].

S5. The calculation method of GRs in NPF events

The growth of newly formed particles for NPF events are often reflected by the collective shift of measured particle size distribution towards larger sizes as time evolves, but it is unfeasible to track the growth of a single particle based on the measurements. Therefore, both approaches used in this study (mode-fitting and appearance time) are referred to as collective approaches and the GRs in this study are the estimated ones (Stolzenburg et al., 2023). While using mode-fitting method, the growth trajectory of new particles is represented by the peak diameter (d_p) of the nucleation mode, which is determined after applying log-normal distributions to the measured size distribution (Kulmala et al., 2012). For measured particle number size distribution at each time (t), there will be a d_p , and the value of GR (GR_{mode}) is derived by a linear fit to the d_p vs t .

Instead of tracking the shift of peak diameter for a given time period, appearance time method seeks to find the time it takes (Δt) for the particle to grow between instrument size bins (Δd_p). In this study, we take the time (t) that the measured concentration of particles reaches its half maximum for each d_p . For each particle size bin (d_p) of the instrument used at BUCT/AHL, there will be a t , and the value of GR (GR_{apt}) is derived by a linear fit to the d_p vs t . Additionally, we believe it would be conceptually more correct to consider diameter as the independent variable when fitting the GRs using appearance time method although previous studies commonly take appearance time as the independent variable and diameter as the dependent variable when fitting the GRs. That is because each data point corresponds to a precise size bin, and any variation (largely stemming from uncertainty due to atmospheric heterogeneity) among the data points primarily exists in appearance time. Consequently, the fitting method with diameter as the independent variable was named as APT-y and the widely used one with time as the independent variable was called as APT-x.

However, it is important to note that the ongoing debate regarding the accuracy between the MOD and APT methods remains unresolved. At present, there is not a definitive conclusion regarding which method is more credible. Both the MOD (mode-fitting) and the APT (appearance time) methods are commonly employed in determining measured GR in NPF events (Kulmala et al., 2012). Recent studies have shown a prevalent use of the MOD method in calculating GR and recognition of particle growth contributor (i.e., H_2SO_4 and OOMs) (Deng et al., 2020b; Qiao et al.,

2021). Hence, in this study's main text, we have utilized the MOD method for comparison with previous research. However, the results from APT-x and APT-y methods are shown in the SI and are reflected in the main text. Hence, in this study's main text, we have utilized the MOD method for comparison with previous research. However, the results from APT-x and APT-y methods are shown in the SI and are reflected in the main text.

It is imperative to view the discrepancies among these methodologies as inherent uncertainties in GR calculations rather than solely attributed to the MOD method itself. For instance, the GRs for sub-3 nm particles determined using APT-x, APT-y, and MODE on May 26, 2021, were 3.17, 5.13, and 2.22 nm h⁻¹, respectively, resulting in an average discrepancy of 1.93 nm h⁻¹ between both APT methods and MODE. However, these values changed to 1.50, 1.92, and 1.93 nm h⁻¹, respectively, on Jun 21, 2021, leading to a comparatively minor deviation. Moreover, the disparity in GRs for 3-7 nm particles across these methods is more substantial as expected, given that the GRs for these particles are consistently more than those of sub-3 nm particles. Our analysis reveals an average discrepancy among those methods of 1.5 nm h⁻¹ for sub-3 nm particles and 3.1 nm h⁻¹ for 3-7 nm particles, reflecting the level of uncertainty associated with GR determination.

(10) Line 586-588: "This is based on the observed consistency between the gaseous H₂SO₄ concentration and its significant contribution to the sub-3 nm particle growth rate in Beijing (Deng et al., 2020b)." However, in this study (Table S4), GR_{1.5-3} was generally higher than GR(H₂SO₄) in Beijing indicating contributions from other species.

Response: Thanks for the comment. Sulfuric acid (H₂SO₄, SA) is one of the most important species in atmospheric nucleation and significantly contributes to particle initial growth (Dunne et al., 2016; Yao et al., 2018; Deng et al., 2020a). The GR(H₂SO₄) is the estimated contribution of SA to particle condensational growth according to Stolzenburg et al. (2020). However, the reviewer is right that other organic materials may make some contribution to the initial growth as well (Salma and Németh, 2019; Mohr et al., 2019; Deng et al., 2020a). Therefore, with respect to our analysis, we remark SA as a significant GR contributor instead of the sole dominant vapor for particle growth in all sizes. Also, the results of SORPES regarding the discussion of SP enhancement are therefore disadvantaged compared to the BUCT/AHL results due to the lack of direct GR measurements. And the revised statements are shown as below,

[Line 295-299] "But it should be noted that H₂SO₄ does not solely dominate the initial growth process, other organic species may make some contribution as well (Stolzenburg et al., 2023). Therefore, in this study, the results calculated at SORPES are disadvantaged compared to the BUCT/AHL data using directly measured GRs. This part of the results should be considered a compromise due to the absence of direct GR measurements."

(11) Fig. 5: In my understanding of the text, although HIO₃ is not the major contributor in the measured GR, the sub 3-nm SP is so sensitive to additional GR

from HIO_3 that the additional HIO_3 enhances SP of sub 3-nm significantly (as shown in fig. 7). And fig. 5 is a nice place to visually indicate that sensitive regimes that correspond to the data in fig. 7.

Response: Thanks for your good suggestion. We have put the field data points in Figure 5 to visually indicate the sensitive regimes of SP to GR. From the following figure (Figure R4), the results demonstrate that minor HIO_3 contribution to GR can result in significant enhancement of particle survival probability, especially in the sensitivity transition regimes for sub-3 nm particles (from blue to red).

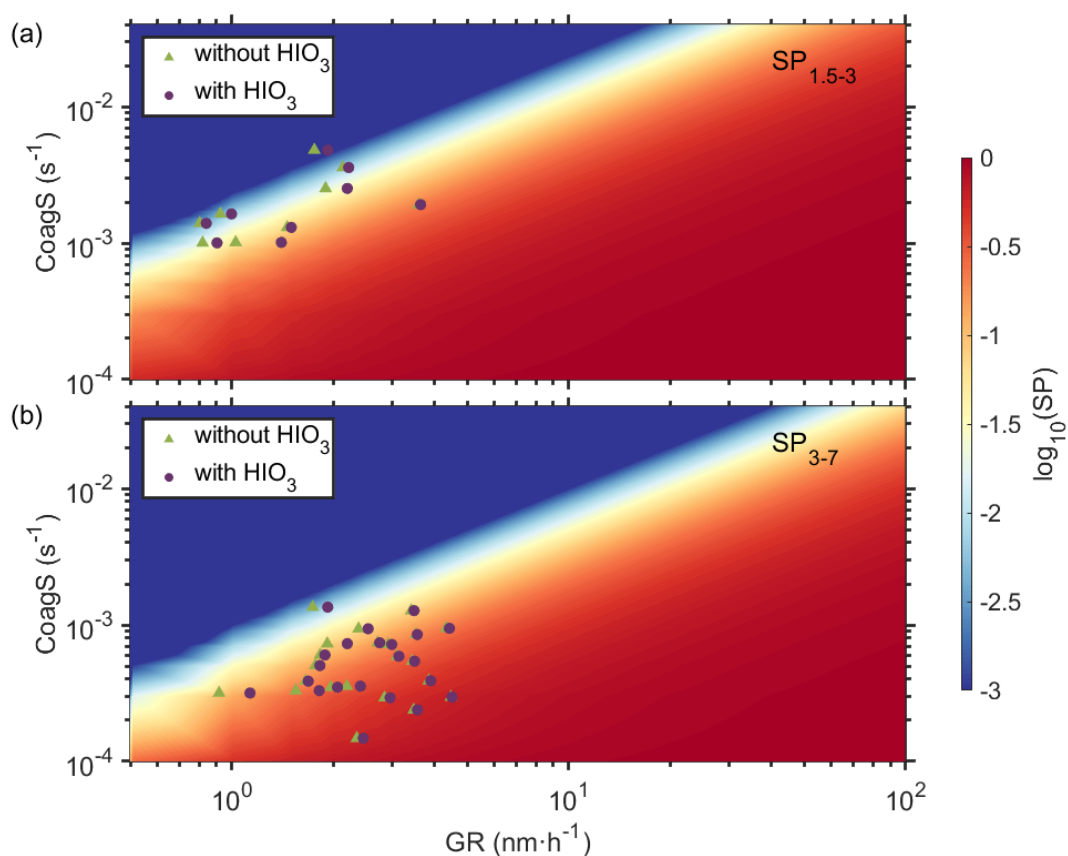


Figure R4. The effect of coagulation sink (CoagS) and growth rate (GR) on particle survival probability for 1.5-3 nm (a) and 3-7 nm (b) particles, respectively. The triangular and circular points represent GR without and with iodic acid contribution, respectively.

(12) Line 606-608: “As depicted in Fig. S8(c), SP enhancements in percentage are generally one order of magnitude higher than the GR contribution in percentage and HIO_3 can result in as high as 2-fold enhancement on SP in sub-3 nm particle growth.” How important is the ~100% (or ~40% on average) enhancement of SP

from HIO₃ in the occurrence of NPF event? In Table S4, during NPF events in Beijing, the survival probability (“P1”) of sub 3-nm particles spans over 3 orders of magnitude from 6.1E-4 to 2.1E-1. Also, that wide range of SP needs to be directly mentioned in the main text.

Response: Thanks for the suggestions. NPF is a crucial process for the Earth’s aerosol system as it is estimated to contribute majorly to aerosol population as well as cloud condensation nuclei (CCN) (Kuang et al., 2009; Gordon et al., 2017). A population of newly formed particles need to grow efficiently to reach typical sizes (usually accumulation or Aitken-mode sizes) in order to be activated to become CCNs, during which they are likely to be scavenged by pre-existing aerosol particles (McFiggans et al., 2006; Kulmala et al., 2001). Hence, survival probability (SP), which characterizes the fraction of freshly formed particles that survive the scavenging process, is an essential and irreplaceable parameter for evaluating the atmospheric influence of NPF. The knowledge of particle SP in ambient atmosphere is limited despite its importance. In this study, we not only reported the theoretical SP calculated using GR and CoagS, and we estimate the influence the potential enhancement of SP with the help of additional GR contributor as well, such as HIO₃. The derived enhancement (up to 100% in some cases) indicate that the fraction of freshly nucleated particles survives the scavenging after a certain growth process is doubled, thus enhancing the atmospheric influences of NPF events. Furthermore, the measurement sites in this study are both located in environments with intensive human impacts where the coagulation sinks are commonly higher. Therefore, the enhancement of SP is even more significant for the new particles to survive the counterbalance of growth process and scavenging by pre-existing aerosols.

Following your suggestions, we described the wide range of SP variations in the revised manuscript.

[Line 573-575]: “It also means that the SP of these newly formed particles exhibits a substantial variability, spanning more than three orders of magnitude, as illustrated in Tables S2~4.”

Technical comments:

(1) Line 94: “survival probability (Kulmala et al., 2017)...” According to the rest of the text, the survival parameter is from Kulmala et al. (2017) and the survival probability is from Lehtinen et al. (2007).

Response: Thanks for the suggestion. We have replaced the reference here.

(2) Line 250: GR'(HIO3) -> GR(HIO3).

Response: Corrected.

(3) Line 269: Define $d_{p_{initial}}$ and $d_{p_{final}}$.

Response: Thanks for the suggestion. We have revised the statement here to be more specific:

[Line 299] “where $d_p = \frac{d_{p_{initial}} + d_{p_{final}}}{2}$ in nm, and the subscripts initial and final refer to the particle diameter at the beginning and the end of the growing process. $[H_2SO_4]$ is the gas phase H_2SO_4 concentration in molecule cm^{-3} .”

(4) Line 594: “IA” and “SA” not defined previously (supposedly HIO3 and H2SO4?). Use consistent names in the main text and SI unless otherwise needed.

Response: We apologize for not being clear. We have checked the relevant expressions to give the consistent names for acids.

(5) Fig. 1&6: Make the panels bigger.

Response: Updated.

(6) Table S8 and S9 caption typo: SOREPS -> SORPES.

Response: Corrected.

(6) Table S2-S7 & S9: Please use the consistent notations for survival probability (SP) and enhancement factor (EF) rather than “P” and “E”. Especially, “P” was used for the survival parameter in the main text and Fig. S6.

Response: Corrected.

(7) Fig S4: The back trajectory color scheme is not consistent. Also, specify the meaning of (%) that the colors represent in the caption.

Response: Thanks for the suggestion. Actually, the color scheme used for the back trajectories is consistent with their respective levels across all four panels. Furthermore, the percentage associated with each trajectory indicates the probability of air mass originating from the corresponding cluster. Also, we have added some detailed discussions about the analysis in the caption of Fig. S5.

And to avoid confusion caused by the color of different clusters, we further revise Fig. S5 as below,

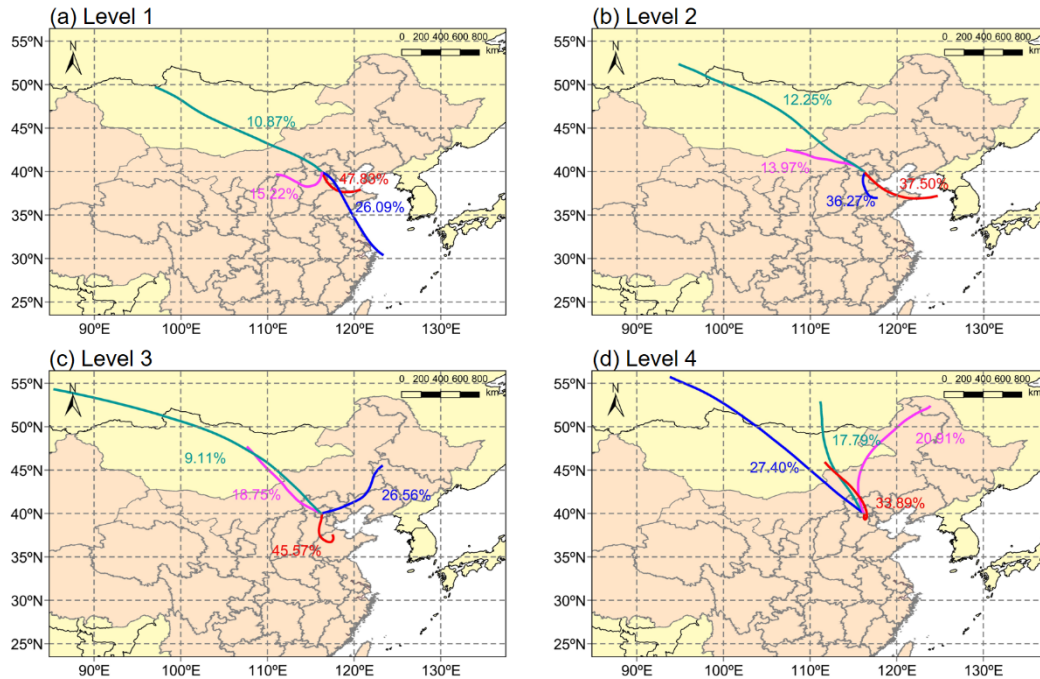


Figure S5. The cluster analysis in different HIO₃ precursors intensities. The four levels of the proxy concentration of HIO₃ precursors are in the 75% - 100%, 50%-75%, 25%-50%, 0-25% percentiles from the first to the fourth levels, respectively. The percentage of each trajectory reflects the ratio of the corresponding cluster.

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