

Response to Reviewer #1

We sincerely thank the reviewer for the valuable feedback and comments, which have helped to improve our manuscript. In the following, we address the general and specific comments point by point. The reviewer's comments are given in ***black bold italic***, while our responses are in normal font (non-bold, non-italic). Revised sections in the manuscript text in response to the comments are written in blue. The page and line numbers refer to the revised version. The figures and tables used in the responses are labeled the same as the revised version.

Comments and suggestions:

In this measurement report, the number size distribution of sub-40 nm particles derived from different instruments in the Amazon rainforest has been reported, including the diurnal pattern and the sources of nanoparticles, especially for the particles below 10 nm. This study provides valuable information in understanding the particle sources in Amazon region and highlight the necessity about the study on nanoparticles. However, further discussion needs to be added to make the results more robust. I recommend this paper being accepted after the below issues are addressed.

Responses and Revisions:

We thank the reviewer for the positive evaluation of our manuscript. We also appreciate the constructive suggestions provided, which have helped us to improve the clarity and robustness of the manuscript. In the revised version, we have carefully addressed all specific comments and incorporated necessary clarifications and additional discussions as recommended.

Comments and suggestions:

Line 45, It is necessary to clearly state that with or without a core sampling device, how much the sampling efficiency was changed for PSM.

Responses and Revisions:

Thanks for the comments. In addition to minimizing the inlet line length to reduce particle loss, we also employed the core sampling device to further improve transmission efficiency for sub-3 nm particles. Figure S1 presents the detailed size-dependent transmission efficiencies for inlet systems with and without core-sampling. Specifically, the transmission efficiencies for 1.5 nm and 3.5 nm particles without the core sampler were 0.62 and 0.85 respectively, whereas with the core sampler, the efficiencies increased to 0.75 and 0.90. These values highlight the improved performance of the inlet system with the core sampler for detecting sub-3 nm particles.

The revised manuscript has included the following clarification sentences (Line 149-151).

“Specifically, the transmission efficiencies for 1.5 nm and 3.5 nm particles were 0.62 and 0.85 without the core sampler, and increased to 0.75 and 0.90 with the core sampler, respectively. This highlights the improved performance of core-sampling in reducing particle loss for sub-3 nm particles.”

The sentence also had been added in the revised Supplementary Material (Line 16-17).

“Figure S1. The transmission efficiency for inlet systems with and without core-sampling. The size-dependent transmission efficiency was calculated with the way introduced by Von der Weiden et al. (2009).”

Comments and suggestions:

Section 2.2.4, is there any calibration was conducted before or after for the intercomparison between nCNC and TSI SMPS? Or the two instruments' inherent system error was lower than 20%? It would be better if the authors can the information about if there are any other studies did such intercomparison, otherwise, how could we know the 20% discrepancy is a "good agreement". In figure 1, please make sure the slope is positive or negative.

Responses and Revisions:

Thanks for the valuable comments. During the intercomparison experiments, we also employed a Grimm Faraday Cup Electrometer (Model 5705, shown in Figure S2) to calibrate and determine the actual cut-off sizes for each instrument. The measured cut-off diameters were 1.7 nm for the TSI system and 1.5 nm for the Airmodus PSM. Given that the resolution of the DMA 3086 is not ideal for such small particle sizes, and considering the overall good agreement observed between the TSI and Airmodus systems, we did not apply further adjustments to the calibration curves or settings of the two instruments.

Regarding the reported ~20% discrepancy, we have summarized relevant intercomparison results from previous studies in Table 1. Similar lab experiments using WO_x particles have shown discrepancies within ±20-35%, and even larger deviations (up to a factor of 10) have been reported in chamber experiments. Thus, we consider the results observed in our intercomparison experiments to be within an acceptable and expected range.

Table 1 Summary of the instrument comparisons (Kangasluoma et al., 2020)

Instruments	Test aerosol	Concentration Range (cm-3) dN/dlogDp	Size range (nm)	Factor between the instruments	Reference
PSM, SMPS	WO ₃	10 ⁴ -10 ⁶	1.2-2.6	<1.2	(Kangasluoma et al., 2015)
PSM, SMPS	WO ₃	10 ³ -10 ⁵	1.5-4	1-1.35	(Cai et al., 2018)
PSM, NAIS, SMPS	Chamber experiment	10 ¹ -10 ⁵	1.3-1000	1-10	(Kangasluoma et al., 2020)
PSM, SMPS	WO ₃	10 ² -10 ⁵	1.8-3.8	1-1.2	This study

We have updated the manuscript accordingly (Line 201-203).

“This level of agreement is consistent with previous intercomparison studies, which reported discrepancies typically within ±20-35% under similar experimental conditions (Cai et al., 2018; Kangasluoma et al., 2015).”

Additionally, we ensured the slope in Figure 1 is clearly indicated and labeled with its correct sign.

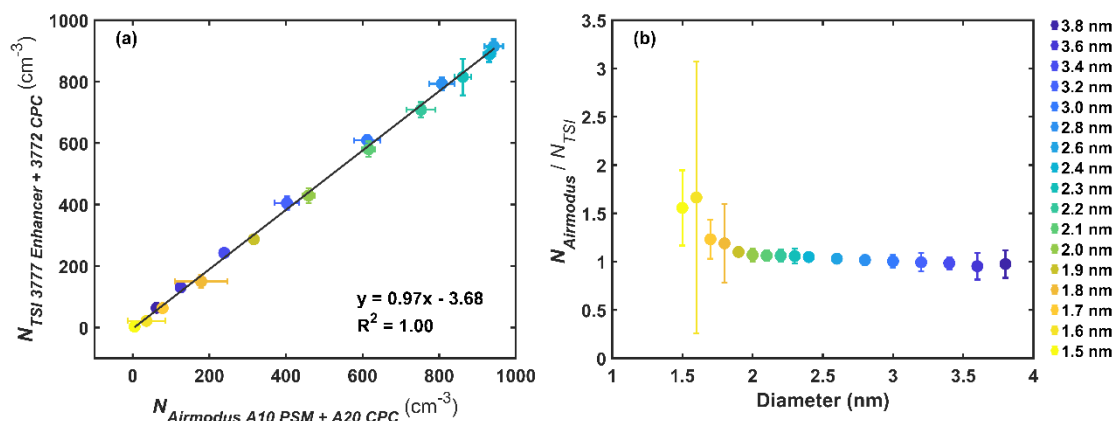


Figure 1. (a) Comparison between the median particle number concentrations measured by two systems, namely the Airmodus (A10 PSM + A20 CPC) and the TSI (Nano-enhancer 3777 + CPC 3772) systems, for the selected different sizes (diameters ranging from 1.5 to 3.8 nm). The solid line represents the linear fitting result with an R^2 value of 1. (b) The ratio of the particle concentration measured by the two systems at different sizes. All error bars denote the standard error.

Comments and suggestions:

Line 210, it should be better to give the definition of “Amazonian bananas” here, otherwise, it may lead the reader to mistakenly think it refers to banana shaped NPF.

Responses and Revisions:

Thanks for pointing this out. We agree that the term “Amazonian bananas” could be misinterpreted as referring to “banana-shaped” new particle formation (NPF) events. However, the term “Amazonian bananas” does not have a formal definition. Unlike typical NPF events, which begin with particle growth from a few nanometers, the “Amazonian bananas” describe growth patterns that typically start at larger sizes, around 20 to 40 nm, as observed in previous studies (Pöhlker et al., 2018; Franco et al., 2022). We have updated the manuscript accordingly (Line 218-220).

“We observed several instances of 'Amazonian bananas', characterized by particle growth initiating at a diameter between 20 and 40 nm, consistent with the observations reported by Pöhlker et al. (2018) and Franco et al. (2022). Notable examples include December 7 and January 20, which are highlighted in Fig. 2c.”

Comments and suggestions:

Line 220-227, is the “high RH range (90-100%)” corresponding to the precipitation? How much is the data excluded from the dataset due to precipitation or high RH? As the mean RH during this measurement was ~85%, that means the probability of the RH exceeding 85% is high.

Responses and Revisions:

The high RH range (90-100%) does not always correspond directly to precipitation events, but it often reflects periods with elevated moisture levels, including pre- and post-rain conditions or fog. However, RH values reaching 100% are typically associated with active precipitation.

In addition to monitoring RH, we also inspected the particle number size distributions for characteristic needle-shaped bursts that are related to raining events. In our analysis, we manually excluded the data only during periods with RH = 100% and/or by visual inspection of the particle spectra when they revealed clear anomalies. Similar to the approach by Wimmer et al. (2018), non-reliable data were identified visually using surface plots of particle size distributions. In total, approximately 22 % of the data, mainly from periods with rain or extremely high relative humidity (RH = 100%), were excluded from further analysis to ensure data quality.

Meanwhile, based on six years of measurements at the ATTO site, the RH in the Amazon typically ranges from 75% to 95% over the course of a day (Franco et al., 2022).

We further clarify this point in the manuscript (Line 236-239).

“Accordingly, all data potentially affected by precipitation were carefully checked and excluded from the NAIS dataset prior to further analysis. This was done by manually identifying periods with RH = 100% and visually inspecting the particle number size distributions for needle-like burst anomalies typically associated with rainfall events.”

Comments and suggestions:

Line 240, have you looked in to the information about radiation and cloud cover in this study? In addition, as the authors have mentioned the high RH during the measurement, it can also be a reason why no typical NPF was observed. As water vapor can also contribute to the high condensation sink in the ambient air, whereas the CS is normally calculated based on the dry PNSD.

Responses and Revisions:

We thank the reviewer’s good suggestion. We agree that factors such as solar radiation, cloud cover, and high relative humidity (RH) can play important roles in impacting new particle formation (NPF) processes. In this study, although we did not include direct measurements of solar radiation or cloud cover, we acknowledge their potential impacts on photochemistry and particle formation. As in Fig. 6 and Fig. 8, we show clearly the sub-3 nm and cluster ions concentration increased as sun arise and peaked around noon, which followed the same trend as the radiation diurnal cycle (Franco et al., 2022). More detailed mechanistic discussions related to photochemistry-induced NPF are out of the scope of this measurement report.

Moreover, we agree that water vapor could potentially contribute to the suppression of typical NPF events by increasing the condensation sink and scavenging nucleation precursors (Andreae et al., 2022; Hamed et al., 2011). However, the mechanism proposed by Hamed et al. (2011) revealed that the reduced H₂SO₄ production was due to lower OH levels at very high relative humidities (>80 %), which may not apply under conditions where nucleation is driven primarily by organic vapors. Laboratory studies by Heinritzi et al. (2020) have shown that changes in humidity do not significantly affect the formation rate of highly oxidized molecules (HOMs) for monoterpene- and isoprene-mixed precursor systems. Additionally, recent measurements in free troposphere indicate that NPF can consistently occur under elevated RH conditions, particularly in the outflow and detrainment layers of convective clouds (Xiao et al., 2023). On the other hand, water vapors even play essential role in cluster formation, forming sulfuric acid and stabilizing other species (Stolzenburg et al., 2023). Overall, these findings highlight the complex and context-dependent nature of water vapor’s influence on nucleation, underscoring the need for more detailed investigations into its role in both particle formation and growth.

We have added the following sentence into the manuscript (Line 259-271).

“On one hand, water vapor may contribute to the suppression of typical NPF events via increasing the condensation sink and scavenging nucleation precursors (Hamed et al., 2011; Andreae et al., 2022). However, the mechanism proposed by Hamed et al. (2011) revealed that the reduced H₂SO₄ production was due to lower OH levels at very high relative humidity (RH >80 %), which may not apply under conditions where nucleation is driven primarily by organic vapors. Laboratory studies by Heinritzi et al. (2020) have shown that changes in humidity do not significantly affect the formation rate of highly oxidized molecules (HOMs) for monoterpene- and isoprene-mixed precursor systems. Additionally, recent measurements in free troposphere indicate that NPF can consistently occur under elevated RH conditions, particularly in the outflow and detrainment layers of convective clouds (Xiao et al., 2023). On the other hand, water vapors even play essential role in cluster formation, forming sulfuric acid and stabilizing other species (Stolzenburg et al., 2023). Overall, these findings highlight the complex and still not fully understood role of water vapor in particle formation and growth, which likely depends on the prevailing chemical regime and meteorological conditions.”

Comments and suggestions:

Line 263-264, is that reasonable that the December data can represent the dry season as the mean RH was approximate 79%? The authors may refer to the previous literature to check the typical RH levels during the dry season in Amazon.

Responses and Revisions:

Thanks for raising this important point. According to long-term measurements at the ATTO site spanning from February 2014 to September 2022, relative humidity (RH) typically ranges from approximately 75-95% throughout the year and 80-100% during the wet season (Franco et al., 2022). Wimmer et al. (2018) reported that the median RH inside the rainforest was 96.9% during the wet season and 94.4% during the dry season. During our measurement period, the average RH was $78.8 \pm 14.4\%$ in December and $89.9 \pm 11.6\%$ in January, indicating that our December RH values were lower than the typical dry season values reported in earlier studies. This indicates that our campaign in December experienced much drier conditions.

Regarding the classification of wet and dry seasons, the studies by Pöhlker et al. (2016) and Andreae et al. (2015) defined the dry season as from August to November and the wet season from February to May. However, Wimmer et al. (2018) followed the definition of Artaxo et al. (2013), in which the dry season extends from July to December, and the wet season from January to June. Thus, we generally think that December and January correspond to the end of the dry season and the beginning of the wet season, respectively. The seasonal context is already clarified in the revised manuscript (Lines 120-121): “Therefore, the measurements carried out in December and January represent the end of the dry season and the beginning of the wet season, respectively.”

Comments and suggestions:

Line 368, it is addressed that on clean days the photochemistry is favorable for the production of nucleated particles. however, it is also stated that there is no typical NPF cases occurred. So that means the burst of nucleation existed, but without clear growth processes. Please give more explanation about the reason.

Responses and Revisions:

Good suggestion. As noted, we did observe signs of nucleation on clean days, indicating that the initial formation of nanoclusters does occur. However, we emphasize that the number concentration of sub-3

nm particles remains relatively low compared to typical NPF events. Moreover, the absence of a clear and sustained growth process suggests that these nucleated particles do not efficiently grow to larger sizes.

We propose two possible explanations for this phenomenon. First, there may be a lack of sufficient low-volatility vapors or precursors necessary to promote particle growth beyond the initial nucleation stage. Alternatively, or additionally, the newly formed particles may be rapidly lost due to coagulation with larger pre-existing particles or through condensation onto surfaces, preventing their survival and growth.

We have added further clarification to the manuscript to reflect these interpretations (Line 402-407).

“However, the number concentration of sub-3 nm particles remained relatively low compared to typical NPF events (Hong et al., 2023; Deng et al., 2022; Bianchi et al., 2016; Kulmala et al., 2013). The absence of clear growth suggests either a lack of condensable vapors to sustain particle growth or efficient loss processes such as coagulation with larger particles (Stolzenburg et al., 2023).”

Comments and suggestions:

Line 405, the first sentence is not rigorous, as the authors also mentioned other studies (such as Wimmer et al., 2018 and Zhou et al., 2002) also conducted the particle size measurement below 10 nm. Please highlight the difference between this work and the others.

Responses and Revisions:

Thanks for the suggestion. We have revised the related text to better highlight the unique aspects of our work. Compared to previous studies such as Wimmer et al. (2018) and Zhou et al. (2002), which conducted measurements near the ground surface, our study presents the first measurements conducted above the forest canopy in the central Amazon, providing data more representative of boundary layer conditions. Additionally, our study is the first to report sub-3 nm particle concentrations and their diurnal variation in this region.

We have revised the sentence accordingly in the manuscript (Line 444-446).

“This study is the first to provide a detailed description of the size distribution and diurnal variation of particles and ions smaller than 3 nm measured above the forest canopy in the central Amazon region, supposed to offer a more accurate representation of boundary layer conditions of the region.”

Table 1, the unit of particle number concentration should be given

Thanks for pointing this out. We have added the unit of particle number concentration in Table 1 as suggested.

Table 1. A summary of the particle number concentration for each instrument in different size ranges. Numbers represent median values, with the 25th-75th percentiles in parentheses.

Instrument	Size range (nm)	Number concentration (cm ⁻³)		
		2022.12	2023.01	All period
PSM*	1.5-1000	963 (679-1363)	972 (775-1287)	969 (735-1320)
	1.5-3.5	371 (251-740)	573 (406-909)	491 (305-851)
	3.5-1000	481 (355-611)	335 (258-410)	380 (295-539)
NAIS (particle)	2-40	1799 (1427-2195)	1141 (883-1489)	1462 (1076-1944)
	2-4	931 (715-1177)	558 (393-778)	749 (509-1032)
	4-12	497 (400-614)	306 (233-401)	403 (290-538)
NAIS (Ion)	0.8-40	769 (677-876)	751 (644-866)	761 (661-872)
	0.8-2	610 (532-712)	639 (543-746)	624 (537-728)
	2-7	40 (34-50)	23 (18-31)	33 (23-43)
	7-20	35 (26-45)	24 (16-35)	30 (20-41)
SMPS	10-40	17 (9-47)	35 (18-73)	27 (13-63)

* PSM refers to the condensation activation diameters (Kelvin equivalent), while the sizes reported by other instruments represent electrical mobility diameters.

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