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Title: Hygroscopic properties of newly formed ultrafine particles at an urban site surrounded by deciduous forest (Sapporo, northern Japan) during the summer of 2011

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Responses to the reviewer's specific comments and questions:

**Reviewer #2 (Comments):**

Jung & Kawamura present results from a hygroscopicity TDMA measuring ultrafine particles in an urban setting in Sapporo, Japan. The focus of the paper is to characterize particles during NPF events, and the authors find that the measured hygroscopicities are in line with particle growth occurring mainly by organic condensation. While the results do merit publication, there are several improvements required for the paper to be publication-ready.

The main problem is that the language is not at an appropriate level, which is clear right from the first sentence of the abstract. This is problematic for the paper's scientific output, as it in some places leaves the reader unable to understand exactly what is meant, and in some leads to apparent claims like that in the abstract stating that wind direction controls hygroscopic properties of particles, which is certainly not the case. In my opinion, the authors should make the effort to have the entire paper proof-read by someone with expertise in atmospheric sciences, as the problems are not always purely grammatical. I found the general structure of the paper to be adequate.

Specific comments

*There were too many grammatical issues that I felt needed correction, so I did not make an attempt to list them all here. I hope the authors can fix them by having the paper proof-read by an expert. Other detailed comments are found below.*

**Response:** The revised manuscript was proof-read by a native English speaker before re-submission.

*$g(RH)$  is defined on page 8262 in Eq. (1), and there it is also mentioned that lognormal modes were fit to the data and mode peak diameters are used in this study. However, it seems to me that the measured "full" growth distributions are used throughout the paper, and the fits are not used except in section 3.4.*

**Response:** We agreed to the reviewer's comment. The phrase "Since a mode peak ~in this study" in P8262, L12-13 in the original MS was deleted.

*Figure captions should provide more details. For example, how are the error/uncertainty bars in Fig. 3, 6 and 7 calculated? What do the vertical lines in Fig. 5 depict? Similar details should be included in all figure captions.*

**Response:** Following sentence has been added in Fig. 4 caption.

"Error bars in (b) represent standard deviations ( $1\sigma$ ) of SO<sub>2</sub>, NO, and O<sub>3</sub> concentrations."

Following sentence has been added in Fig. 7 caption.

"Rectangles represent northwesterly or northeasterly wind directions."

Following sentence has been added in Fig. 8 caption.

"Error bar represents  $1\sigma$  of g(85%) value."

Following sentence has been added in Fig. 9 caption.

"Error bars represent  $1\sigma$  of number concentration of less-soluble particles at dry D<sub>p</sub> = 100 nm and NO concentration."

*Page 8261, line 12: Enriched compared to what? Or do the authors just mean "abundant"?*

**Response:** The term "enriched" in P8261, L12 has been changed to "abundant". Please see line 113 in the revised MS.

*8262, 12-14: What does this sentence mean? Both DMAs will have transfer functions, but what transfer function between the DMAs are the authors talking about?*

**Response:** We agreed to the reviewer's comment. The phrase "broadening ~negligible." in P8262, L13-14 in the original MS was deleted.

*8262, 24: Presumably wind direction was also measured as it is discussed. What about the NO and ozone measurements?*

**Response:** The NO and O<sub>3</sub> measurements were already mentioned in P8260, L23-24 in the original MS. The term "wind direction" has been added in line 150 in the revised MS.

*8264, 4: "With and without a timegap" is not clear. Please explain.*

**Response:** The sentence starting "Increases in the number..." in P8264, L2-4 in the original MS was modified as follows.

"Increases in the number concentrations of humidified particles at a dry D<sub>p</sub> of 40 nm were observed after the burst of humidified particles at a dry D<sub>p</sub> of 20 nm occurred."

Please see lines 201-203 in the revised MS.

8265, 4: How is this  $g(85\%)_{total}$  defined? I doubt the “\_” is necessary here.

**Response:** We agreed to the reviewer’s comment. The term “ $g(85\%)_{total}$ ” in sections 3.2 and 3.6 were changed to “ $g(85\%)$ ”.

8265, 20-21: Then this should show a correlation with increased particle number in each size bin. Is this the case? Otherwise, some other explanation is needed. Perhaps less volatile condensable vapors are emitted that can condense on existing particles? The larger question is why this effect is not seen during non-npf days? Is it due to different air masses leading to npf vs non-npf days, or are npf days typically sunny days where vertical mixing is more efficient in the mornings?

**Response:** We agree to the reviewer’s comment. Following sentences have been added in lines 258-260 in the revised MS.

“As seen in Fig. 5,  $g(85\%)$  decreases with an increase in the number concentration of particles in each size bin between 4:00 and 8:00 LT.”

Following figures have added in Fig. 5 in the revised MS.

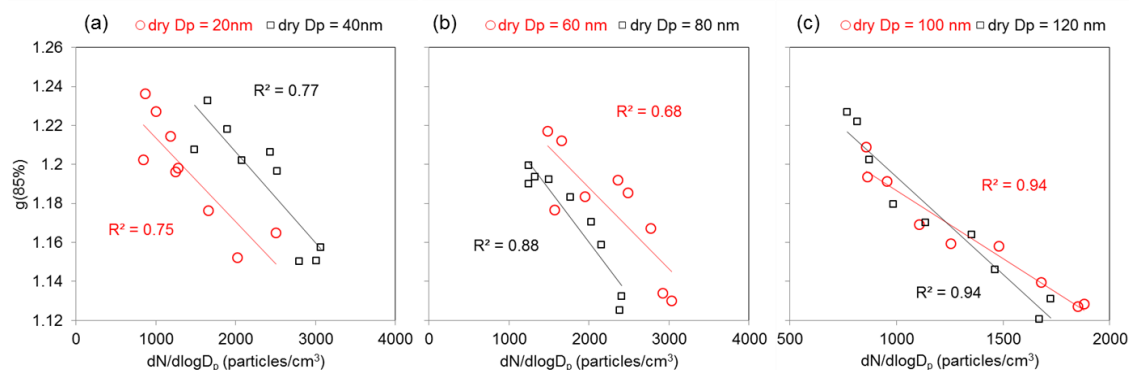


Fig. 5 Scatter plots of  $g(85\%)$  values versus particle number concentrations in each size bin during the NPF event days. (a) dry  $D_p = 20$  nm and 40 nm, (b) dry  $D_p = 60$  nm and 80 nm, (c) dry  $D_p = 100$  nm and 120 nm. The data points are 30 min averaged  $g(85\%)$  values and particle number concentrations from 4 A.M. to 8 A.M. marked (A) in Fig. 4a and c.

8268, 3 & Table 1: “Grown Aitken mode nucleated particles” is not a good term. The authors can define that nucleated particles that have grown to Aitken mode sizes are studied, and then just refer to the Aitken mode particles.

**Response:** We agree to the reviewer’s comment. The phrase “grown Aitken mode nucleated particles” in P8264, L5 has been changed to “nucleated particles that had grown to Aitken mode sizes” in lines 204 in the revised MS.

The term “grown Aitken mode nucleated particles” in section 3.4 have been changed to “the

Aitken mode particles” in the revised MS.

8268, 26: "Aitken".

**Response:** Corrected.

8269, 20: *The authors should look over how many times “accounting for the Kelvin effect” is mentioned in the paper. It is enough to define it once and then use the Equiv.g(RH).*

**Response:** We agree to the reviewer’s comment. The sentence starting “g(85%) at dry  $D_p = 20\text{nm}...$ ” in P8266, L22-24 and Fig. 6 caption was changed as follows. Please lines 283-284 in the revised MS.

“g(85%) at dry  $D_p = 20\text{ nm}$  in Fig. 6 is Equiv. g(85%) at dry  $D_p = 100\text{ nm}.$ ”

The sentences starting “g(85%)...” in P8269, L19-20 and Fig. 5 caption in the original MS were deleted.

8270, 10-12 (and earlier): *Such a conclusion is not correct. The observed g(RH) at the station is perturbed by the wind direction, by bringing different air masses to the station. The actual growth of the particles will depend on other factors, and is thereby something that cannot be captured solely by the data obtained at this station.*

**Response:** We agree to the reviewer’s comment. The sentences in P8270, L10-12 in the original MS have been modified as follows. Please lines 393-395 in the revised MS.

“Thus, the hygroscopic growth factor of newly formed particles was perturbed by the local winds that delivered different air masses to the measurement site.”

8271, 15-16: *”In contrast, similar figures were obtained for g(85 %)\_more in both periods as shown in Fig. 7.” What is the importance of this parameter. As I understand it, it is the average growth factor of particles with growth factors larger than 1.25, which sounds like a relatively artificial number. If there is scientific information in this number, the authors should explain it.*

**Response:** We agree to the reviewer’s comment. g(85%)\_more in Fig. 7 in the original MS was deleted. The sentence “In contrast, ... in Fig. 7” in P8271, L15-16 in the original MS was deleted.

*Fig. 1. I am happy to see a map of the surrounding area, but I still have no idea where the measurement site is located. There are two red boxes added, but neither is described. More details are needed here.*

**Response:** As suggested by the reviewer, Fig. 1 was slightly modified to clearly see the measurement site. Figure 1 caption was also modified as follows.

“Fig. 1 Map showing the measurement site (red rectangle). The measurement site is located on the north campus of Hokkaido University (43°3'56" N, 141°21'27" E) in the northwest of downtown Sapporo, northern Japan. Observed frequencies of local wind direction with wind speed are also shown.”

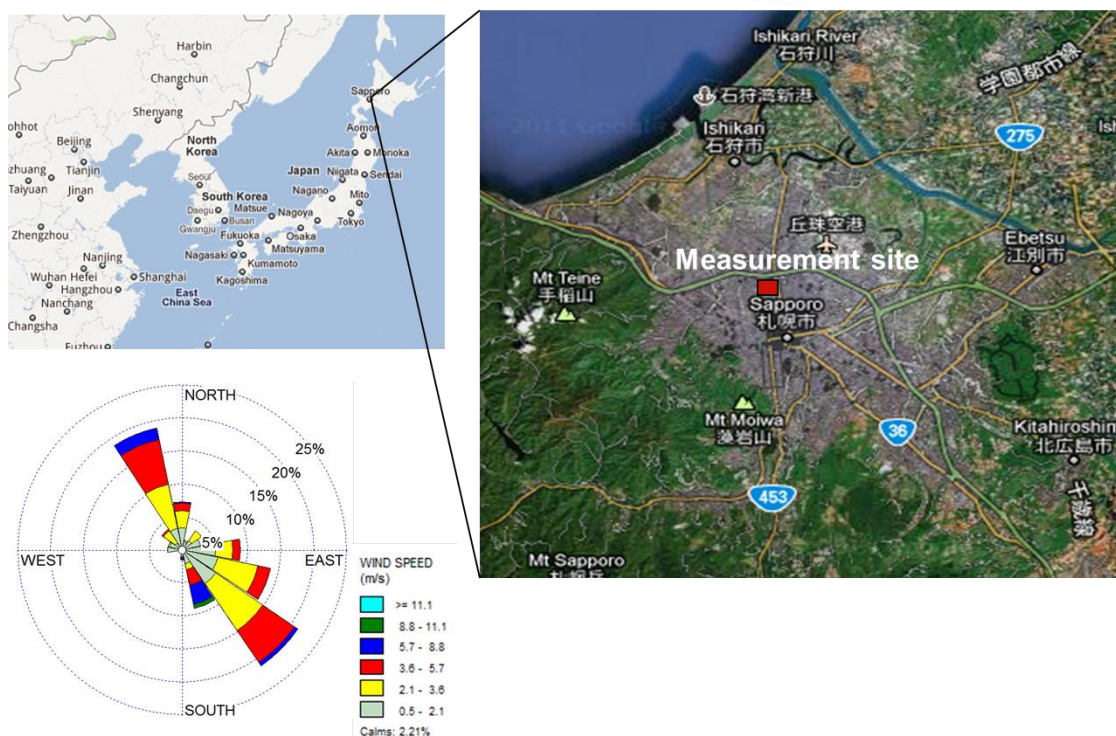


Fig. 2. Panel (a) should be referenced and discussed first.

**Response:** As suggested by the reviewer, panel (a) in Fig. 3 in the revised MS was referenced and discussed first.

Figure 3 caption was modified as follows.

“Fig. 3 (a)  $N_{nuc}$  and (b-g) temporal evolutions of the number concentrations of atmospheric particles as a function of the hygroscopic growth factor at 85% RH [ $g(85\%)$ ] at the measurement site between 27 July and 8 August 2011. Dry particle diameter ( $D_p$ ) increases from (b) 20 nm to (g) 120 nm with a 20 nm increment. Eight new particle formation (NPF) events were identified on 27 and 31 July, and 1–3, 5, 6, and 8 August 2011, as marked by white squares.”

The sentences in P8263, L20-24 in the original MS have been modified as follows. Please see lines 194-197 in the revised MS.

“Figure 3 shows  $N_{nuc}$  and the number distributions of humidified particles at the dry  $D_p$  range of

20–120 nm as a function of  $g(85\%)$ . During the measurement period, eight NPF events occurred on 27, 31 July, and 1–3, 5, 6, and 8 August, as shown in Fig. 3a, and marked as white boxes.”