

Interactive comment on “Observations of Particles at their Formation Sizes in Beijing, China” by Rohan Jayaratne et al.

Response to Comments from Anonymous Referee #1

Overall Comments

This paper contributes to the understanding of some of the factors that control new particle formation (NPF) events in more-polluted regions of the atmosphere. The paper is well written and falls within the scope of the journal. I find the comparisons between NPF and non-NPF days to be of particular interest, along with the detailed measurements of NPF events in the ~2-10 nm range by the NAIS, a range not well-captured by studies that rely on SMPS-type particle number concentration measurements alone. I recommend this paper to be published in ACP with minor revisions, as discussed below.

Response to Overall Comments

We thank the reviewer for these positive comments and are glad to note that the paper falls within the scope of the journal.

General comments:

Comment 1

Page 4, lines 81-83: This statement is confusing. Did the PNC exceed 10^5 cm^{-3} on all 45 days or just for the 25 days that NPF was observed? Please clarify.

Response 1

We accept that this sentence is confusing. We will amend it as follows:

“They observed NPF on 25 out of 45 days of measurement, and on each of these days the PNC exceeded 10^5 cm^{-3} .”

Comment 2

Page 8, calculation of the diffusion coefficient: It would be good to include a brief discussion of the assumption that the main condensing vapor is sulfuric acid. Particle composition measurements were not a part of this work, but the authors do cite Yue et al. (2010) as showing that some NPF events in Beijing had sulfuric acid accounting for much less than half of the total growth rate, with organics accounting for ~55% of the growth. In the kinetic regime, the RMS speed of a molecule depends on $1/\sqrt{\text{MW}}$, where MW = molecular weight = 98 g/mol for sulfuric acid = ~200 g/mol for organics. This would mean that organics would be about ~30% slower, and condensation in the kinetic regime is proportional to RMS speed. The continuum regime is trickier as it depends on the diffusion coefficient instead of

RMS speed, but if we simplify to assume everything is in the kinetic regime, then the CS would scale as $\sqrt{\text{MW of sulfuric acid}} / \sqrt{\text{MW of orgs}} \sim \sqrt{98} / \sqrt{200}$. There are of course limited calculations and measurements of the diffusion coefficients of organic molecules as a function of temperature; however the authors could briefly comment on some of the literature values compared to their assumed value of D using sulfuric acid.

Response 2

In response to these comments, we will incorporate some or all of the following discussion into the paper:

It is now well established that sulfuric acid is the key precursor gas in nucleation, although low vapour pressure organics may contribute to the subsequent aerosol growth (Curtius, 2006). Sulfuric acid has a low vapour pressure which is reduced further in the presence of water. When produced from SO₂ in the gas phase, it is easily supersaturated and begins to condense. Moreover, most of the particles in the atmosphere are in the kinetic regime (smaller than 0.01 μm)(Seinfeld and Pandis, 2006). In this regime, condensation is directly proportional to the RMS speed of the molecules. The RMS speed is inversely proportional to the square root of the molecular weight of the molecule. Thus, a sulfuric acid molecule, with a molecular weight of 98 g mol⁻¹, has an RMS speed that is about 30% higher than a typical organic gas molecule with a molecular weight of about 200 g mol⁻¹. Thus, condensation of sulfuric acid will occur much more readily than organic molecules. Studies in Beijing have confirmed that NPF is more likely to occur in a sulfur-rich environment than in one that is sulfur-poor ((Yue et al., 2010;Guo et al., 2014;Wu et al., 2007)). Wu et al. (2007) also assumed that sulfuric acid was the main condensable vapour in determining the particle formation rates during NPF events in Beijing.

Our estimated values of D for sulfuric acid using the equation given in Jeong (2009) are 0.092 cm² s⁻¹ at 303K and 0.087 cm² s⁻¹ at 273K. The value of 0.092 cm² s⁻¹ at 303K is reasonable as it is similar to other values given in the literature at room temperature, for example Brus et al. (2016) (0.08 cm² s⁻¹) and Eisele and Hanson (2000) (0.095 cm² s⁻¹). The values of D for common organic trace gases as given in the literature are somewhat smaller than this, e.g. 0.07 cm² s⁻¹ for isoprene (Tang et al., 2015) and terpenes (Williams, 2004). D for atmospheric amines are of the same order as that for sulfuric acid (Lugg, 1968) Therefore, we feel that D = 0.092 cm² s⁻¹ is a reasonable value to use in calculating the CS.

Comment 3

Page 9, condensation sink (CS) calculations: Why did the authors (1) choose to use 303 K in their diffusion coefficient (D) calculation (line 204) and (2) only use the SMPS PNC for the CS (line 203)? In regards to (1): temperature data wasn't reported in this paper but was taken as part of the meteorological data. The historical data reports Beijing's average temperature in January as being around ~270 K. This difference in temperature doesn't lead to a particularly large change in D but certainly is worth addressing.

Response 3

3 (1) We accept the point about the temperature. The average temperature in Beijing during the observations was close to 273K. We have re-calculated our parameters using this value for T. The revised values are given below, and these will replace the values in the paper.

3 (2) Regarding the point about calculating the CS, our response to this comment is included in Response 4 below.

Comment 4

In regards to (2): I would like to know why the authors chose to neglect the PNC data obtained from the NAIS for <14 nm size bins. A few calculations with “toy” size distributions show that, depending on the number concentration at these <14 nm bins, the CS can be non-trivially changed with the inclusion of these smaller bins. If the size distributions during NPF events in this paper such that the CS hardly changes with the inclusion of the smaller size bins, this should be stated.

Response 4

We re-calculated the parameters, first by holding the temperature at 303K, in order to check if including the particles smaller than 14 nm will make a significant difference to the CS. We found that the CS value increased by about 8% (from 4.8 to 5.2 s⁻¹).

Therefore, we have re-calculated all the parameters using the value of CS obtained across the entire size range (< 14 nm from the NAIS plus >14 nm from the SMPS) and with the temperature changed from 303K to 273K.

The original values in the paper are as follows:

$$\begin{aligned}D &= 0.092 \text{ cm}^2 \text{ s}^{-1} \\CS &= 4.8 \times 10^{-3} \text{ s}^{-1} \\Coag &= 8.3 \times 10^{-4} \text{ s}^{-1} \\FR &= 23 \text{ cm}^{-3} \text{ s}^{-1}\end{aligned}$$

The new values obtained are as follows:

$$\begin{aligned}D &= 0.087 \text{ cm}^2 \text{ s}^{-1} \\CS &= 4.2 \times 10^{-3} \text{ s}^{-1} \\Coag &= 7.2 \times 10^{-4} \text{ s}^{-1} \\FR &= 26 \text{ cm}^{-3} \text{ s}^{-1}\end{aligned}$$

We will correct the paper accordingly.

Comment 5

Page 13, lines 294-296: It is also worth mentioning that the pre-existing particles coming into the region from the winds from the south are also increasing the condensation sink, further reducing the likelihood of NPF.

Response 5

We will include the following text.

“Pre-existing particles entering the region with the winds from the south will also increase the condensation sink, further reducing the likelihood of NPF.”

Figures/Tables:

Comment 6

Each figure (excepting Fig 4) could benefit from being more professionally presented. I'm not sure what programming language was used to create these figures but if it is e.g. python, using `savefig('name.png',dpi=300)` and `savefig('name.pdf')` would create much nicer looking figures. The text is somewhat blurry and could benefit from being saved at a higher dpi (for png) or as a pdf without the grey backgrounds.

Response 6

Since first submission to ACP required embedding the figures within the body of the manuscript, the resolution of the figures has suffered. In the final submission, we shall present each figure as a separate file, so that they will be of much higher resolution. Also, we will remove the grey background and the frames from all the figures.

Comment 7

Figures 2 and 7: The colorbars need labels of units. The numbers on the colorbars are quite blurry and need to be sharpened.

Response 7

The numbers that appear on the color bars are produced by the software. As we have provided two labels with the end point values, we feel that these intermediate numbers on the color bars are not essential. To compensate for this, we will further sharpen the text labels at the two ends of the color bars.

We will add the units (cm^{-3}).

Technical comments:

Comment 8

Abstract, lines 29-31: The sentence would read better is if said ‘Estimated characteristics... are very different than to when the measurements. . . .

Response 8

The text will be changed as requested.

Comment 9

Page 3, line 57: environments (needs an 's')

Response 9

The "s" will be added.

Comment 10

Page 11, line 247: no comma after 'that'.

Response 10

The comma will be deleted.

Comment 11

Page 16, line 385: This sentence might read better if it said '...in the smallest particle size bin 2-3 nm for the times at which the rate of increase. . .'

Response 11

We agree. The text will be changed as requested.

References:

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