C1563:

General:

In this paper the authors study New Particle Formation events (NPF) in Beijing during the CAREBeijing-2008 campaign in summer 2008. In all 12 NPF events are studied, and particle formation rate and particle growth are investigated. Both observations and modeling are used to analyze particle formation and growth. The role of sulphuric acid is discussed.

This is a well written paper that deserves publication. It is interesting and reveals new aspects of particle formation and growth in the Beijing area. However I have some comments listed below that I think need to be addressed before publication.

Response: Thank you very much for your comments.

Major comments:

1) A key question here is what kind of processes controls the NPF events. Is it sulphur (sulphuric acid), organic compounds or other processes not discussed here? In fig 2a FR is proportional to H_2SO_4. This indicates that sulphuric acid is the "trigger" for this process. Fig. 1 shows the concentration of H_2SO_4 and small particles, both show a maximum around 11a.m. And observations show that concentrations of sulphuric acid are higher during NPF events than during days of non-NPF. All of this supports the idea that sulphuric acid controls NPF. However, in fig. 4b, right panel ("sulphur-poor") there are very low concentrations of smaller particles. Most abundant particle size is around 10-20 nm (at noon). This "absence" of smaller particles should be investigated further. See also comment 5) below.

Response: Thank you very much for your suggestion.

On 31 August the sulfur-rich NPF event happened with high speed wind from the north and the concentrations of isoprene was low (below 1 ppbv), while on 18 September the sulfur-rich NPF event occurred with low speed wind from the north and the concentrations of isoprene kept above 1 ppbv as shown in Figure I. These different conditions might lead to different properties of the NPF events.

For the absence of the smaller particles at the start of the NPF event on 18 September we think possible reasons are: (1) Transport. Some of the nucleation mode particles are probably transported from the surrounding areas to the PKU site during the start of this NPF event on 18 September. During the transport they keep growing and lead to the absence of the very small particles. (2) Fast growth. More pre-existing larger particles were observed before and during the start of the NPF event on 18 September than on 31 August as shown in Fig.4. The newly formed particles are easy to coagulate to the large particles as well as self-coagulate and led to the fast apparent growth rate of the nucleation mode particles (6 nmh^{-1}).

We will add a paragraph about the absence of the smaller particles during this NPF event in the revised manuscript as follows:

Page 2722 before line 6:

At the start of the NPF event on 18 September, the nucleation mode particles were observed to grow from above 10 nm (geometric mean diameter) not as on 31 August from sub-10 nm. Transport of particles from the surrounding areas to the PKU site and very fast growth of the small particles during the very start of this NPF event on 18 September are possible reasons for that and need further investigation.

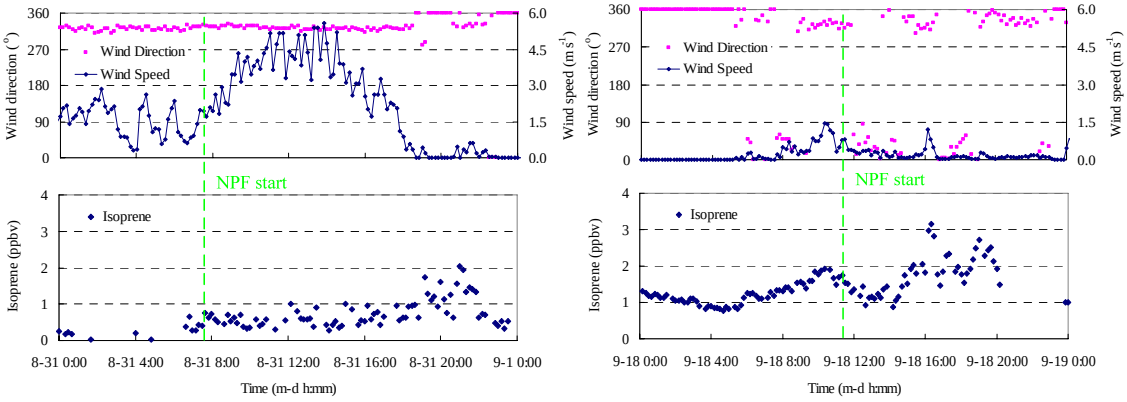

Figure I: Wind speed, wind direction, and concentrations of isoprene on 31 August with a sulfur-rich NPF event and on 18 September with a sulfur-poor NPF event

2) The CAREBeijing campaign took place from 12. July to 25. Septembre 2008. Beijing hosted the Summer Olympics from 8. to 24. August this year. To improve air

quality in Beijing many restrictions were imposed on industry and on traffic in Beijing and surrounding areas. This implies that the chemical composition of the air in Beijing was different just before and during the Olympics, compared to "normal" conditions. 4 out the 12 NPF events occurred during the Olympics (Fig. 1). This means that 1/3 of the events occurred during these 16 days which covers 16/76 (~1/5) of the campaign period. This may indicate that the "cleaner" air played a role. This is not mentioned and should be discussed by the authors.

Response: Thank you very much for your suggestion.

On one hand the pre-existing particles and nucleation process compete for the precursors such as sulfuric acid, which also condense onto the pre-existing particles. On the other hand, the pre-existing particles coagulate the newly formed particles. Therefore, "cleaner" air with less pre-existing particles played an important role in the occurrence of NPF events in Beijing. During the Olympics with the restrictions imposed on industry and traffic, lower loading of particles and higher occurrence frequency of NPF events in the urban area of Beijing was observed.

Discussion about this fact will be depicted in the revised version as follows:

Page 2719 before line 18:

Low condensational sink (corresponding to low particle surface areas concentration) is one of the key factors for the NPF event occurrence in Beijing (Wu et al., 2007). On one hand the pre-existing particles and nucleation process compete for the precursors such as sulfuric acid, which also condense onto the pre-existing particles. On the other hand, the pre-existing particles coagulate the newly formed particles. Therefore, "cleaner" air with less pre-existing particles plays an important role in the occurrence of NPF events in Beijing. During the Olympics with the restrictions imposed on industry and traffic, lower loadings of particles in the urban area of Beijing were observed. That may be one important reason for the fact that the occurrence frequency of NPF events during this period (25%) is about twice of that before and after Olympics (about 13%).

3) The observations were sampled at Peking University, in the N-W part of Beijing.

The paper states that "The NPF events usually occur in conjunction with high wind speed from the north, low relatively humidity (below 45%), and intense solar radiation" (section 3, p. 2718). During episodes with wind from the North I would expect a considerable amount of mineral dust. However mineral dust particles are usually large particles (~μm). Was there any mineral dust present during the NPF events? And if so, how may this mineral dust influence the results and findings in this paper?

Response: Thank you very much for your comment.

High speed wind from the north in the spring can bring much mineral dust to Beijing and cause dust storms sometimes. Usually, the dust events observed in Beijing are associated with cold air advection behind a cold front. This leads to dry and pristine atmospheric conditions, which favors new particle formation (Wu et al., 2009). But in the summer with good vegetation coverage, less mineral dust are presented. On the days with or without NPF events in the summer no obvious difference of coarse particles was observed. Besides, the number and surface area concentrations of the coarse particles are quite low, so they will not impose obvious effect on the NPF events.

4) 7 out of 12 events are characterized as "sulphur-rich" and 5 as "sulphur-poor". In the sulphur-rich events there was 30% higher concentration of sulphate than organic compounds whereas for the sulphur-poor there was 20% lower concentration of sulphate compared to organic compounds. During NPF events the concentration of sulphuric acid was relatively high (5×10^6 cm^{-3}) compared to non-NPF days (typical concentration 2×10^6 cm^{-3}). In figure 3b it seems that there is hardly any difference in sulphuric acid concentration for sulphur-rich and sulphur-poor events. Hence sulphur concentration is almost identical, whereas the sulphur/organic ratio varies, due to changes in organic compounds. To me it would perhaps be more appropriate to call it "organic-poor" rather than "sulphur-rich" and vice versa. Is there any information available about the concentration and composition of organic compounds? The authors should discuss this.

Response: Thank you very much for your suggestion.

But we are sorry that we did not measure the potential organic precursors of NPF such as pinic acid and ketopinic acid and the organic species in the particles especially the ultrafine particles. So the role of organics in the NPF events in Beijing can not be decided. Besides, nitrate may also contribute to the growth of the nucleation mode particles though not much (<10%). In addition, sulphuric acid is the "trigger" and controls the occurrence of the NPF events in Beijing and the fraction of the contribution of sulfuric acid condensation and neutralization to the particle growth is distinct in the two different types of NPF events. So they are called "sulphur-rich" and "sulphur-poor" according to the available data.

5) From figures 1 and 4 it seems that high concentrations of the very small particles occur early during the day (~10a.m. for 12. August in fig. 1 and 8.a.m for 31. August in fig 4). The two lowest panels in fig. 4 show sulphuric acid and organic compounds for events, but only for 2 p.m. to 6.p.m. It would be interesting to see sulphuric acid and organic compounds early in the morning, i.e. when the concentrations of smaller particles are at a maximum, and NPF events start. This would tell us whether organic compounds play a role in the very start of NPF events.

Response: Thank you.

It would be very interesting and useful to do so, but it is a pity that the potential organic precursors of NPF such as pinic acid and ketopinic acid and the organic species in the ultrafine particles have not been measured. The AMS measured the organic matters in particles from 40 to 400 nm, meaning that it can not detect the chemical compositions of the nucleation mode particles which are more critical to study the very start of NPF events. In addition, AMS need a proper mass loading (about 0.4 μgm^{-3} for organic matter and 0.1 μgm^{-3} for sulfate) to guarantee the data veracity. That's one reason why the time period from 14:00 to 18:00 was picked out for the discussion. Further investigation about the role of organic compounds in the early stage of NPF events is necessary with the potential organic precursors and the chemical compositions in the very small particles, for instance 3-10 nm.

Minor comments:

1) Fig. 4: The scales for e and f as well as g and h are different. The left panels go up to 3, whereas the right ones go up to 6. This may be a bit confusing since the colours are the same.

Response: Thank you very much for your comment.

In the revised version we will set the corresponding scales the same to make them more clear and comparable.

Here is the new version of Fig. 4:

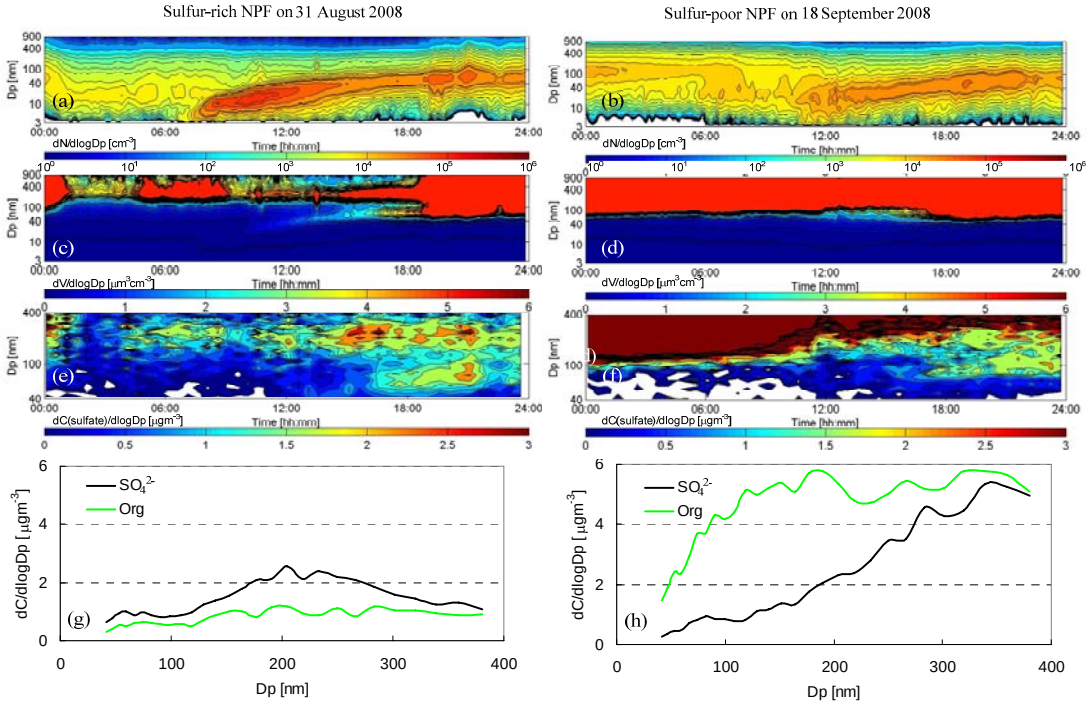

Fig. 4 Particle number size distributions (a, b), particle volume size distributions (c, d), sulfate mass size distributions (e, f), and comparison of average mass size distributions between sulfate and organic compounds from 14:00 to 18:00 LT (g, h) on sulfur-rich and sulfur-poor NPF event days, respectively.

2) In formula (1) FR (formation rate) = dN/dt (change in concentration of N) + F_{coag} + F_{growth}. Here F_{coag} and F_{growth} represent a loss. It would appear more clearly that these are loss terms if there were "minus" rather than "plus". As the formula (2) is written now, Ki (Brownian coagulation coefficient) must be negative since F_{coag} is negative, and Ni is definitely positive. However, if "minus" is used to show a loss, Ki must be positive.

Response: Thank you.

As it is quite complicated and not necessary to discuss the “minus” or “plus” value for the parameters in this paper, the absolute values of them are used in stead.

References:

Wu, Z., Hu, M., Liu, S., Wehner, B., Bauer, S., Mäßling, A., Wiedensohler, A., Petäjä, T., Dal Maso, M., Kulmala, M.: New particle formation in Beijing, China: Statistical analysis of a 1-year data set, J. Geophys. Res., 112(D09209), doi:10.1029/2006JD007406, 2007.

Wu, Z. J., Cheng, Y. F., Hu, M., Wehner, B., Sugimoto, N., and Wiedensohler, A.: Dust events in Beijing, China (2004–2006): comparison of ground-based measurements with columnar integrated observations, Atmos. Chem. Phys., 9, 6915-6932, 2009.