Anonymous Referee #2

We thank this reviewer for helpful comments and suggestions. Below we provide a point-by-point response to this referee's comment. The page numbers refer to the original version published in ACPD.

Comment

In terms of cloud condensation nuclei (CCN), the authors should more clearly point out the existence of the following three different sources: 1) primary particles able to act as CCN, 2) CCN resulting from NPF and subsequent particle growth, 3) primary particles that cannot originally act as CCN but become CCN after ageing (growth and addition of hygroscopic material) in the atmosphere. In analyses as the one performed in the paper, it is extremely difficult, sometimes impossible, to distinguish sources 2 and 3. In practice, this means that the reported some unknown fraction of the reported CCN enhancements due to NPF is, in fact, due to ageing of primary particles. The authors should bring up this issue explicitly in the paper and discuss it shortly.

Response

Thanks your comment. We think it is very important to clarify this in the revised version:

"It also should be clarified that we might produce several potential misunderstanding on estimating the contributions of NPF to CCN number concentration. The CCN in the ambient atmosphere could be formed in the following three ways: 1) primary particles able to act as CCN directly; 2) CCN resulting from NPF and subsequent growth; 3) primary particles that cannot originally act as CCN but become CCN after aging (growth and addition of hygroscopic material). However, it is extremely difficult to distinguish the contributions of diverse sources to the CCN enhancements, particularly which we assumed that all due to NPF, in fact, might also be contributed by the primary emission or the following aging process, especially at the polluted urban environment with the intensive primary emissions by anthropogenic activity. Nevertheless, these effects could be neglected at a regional background site such as

SDZ. Hence, in this section, we only pay attention to the regional NPF events observed concurrently at both sites (total 34 events in Class I type). The potential contributions of regional NPF event to production of CCN are firstly calculated for the regional and urban sites, and the gap is roughly estimated as mainly contributed by the anthropogenic emissions at polluted urban environment. In addition, the regional transportation between these two sites was not evaluated in quantity in this study, which might also cause uncertainties."

Comment

In terms of aerosol light scattering enhancement due to NPF, I am even more skeptical that particles formed originally in the atmosphere would make a dominant contribution in areas influenced heavily by anthropogenic sources. My main argument here is that particles giving the dominant contribution to the light scattering coefficient tend to be larger than 200-300 in diameter and particle formed in the atmosphere do not usually grow that large. The fact that increased light scattering follows NPF could simply be due to more active secondary aerosol formation that involves existing primary particles. The authors should check out carefully which size range really contributes to light scattering and whether newly-formed particles are really able to reach this size range. In case they do not, the authors can simply reformulate their findings by stating something like "aerosol light scattering was increased as a result of secondary aerosol formation following NPF".

Response

We have calculated the particle light scattering distributions, which could reflect the contributions by the diverse modes. The results showed that the particle scattering coefficients were mainly contributed by the accumulation mode particles (> 100 nm). And these newly formed particle could not grow too such a large size. However, the distinct increasing of scattering coefficient due to the particle nucleation was also observed. We have stated these in the revised manuscript:

"In order to demonstrate the influence of the nucleation events, we also calculated the size segregated light scattering distributions at both sites (Figure 5c & 5d). In theory,

the light scattering of a 100 nm particle at wavelength of 550 nm is about 2-order and 3-order of magnitude smaller than those for a particle with diameter of 200 nm and 400-500 nm, respectively. This means in general the light scattering is dominated by relatively large accumulation mode particles (> 100 nm), which was also the case at PKU and SDZ in the beginning of nucleation event (e.g., contribution to the light scattering by particles smaller than 100 nm was always smaller than 1%). Note that it is hard to rule out the contribution of decreasing boundary layer height in the late afternoon and evening, which also leads to the increase in particle mass concentration and therefore light scattering. However, in Figure 5a & 6b, one could clearly see the continuous growth in particle diameters at both sites from nucleation mode (~ 10 nm) in the late morning (10:00, local time) through the Aitken and accumulation modes before the clearly dropping down of boundary layer height (e.g., 16:00, LT). Because of that, the scattering coefficient due to the particle nucleation and subsequent condensable growth kept increasing (Figs. 5c&5d). Although the contribution to total particle light scattering by ultrafine particles (Dp < 100 nm) is still negligible (< 1%) and for particles larger than 100 nm their condensational growth become very slow in terms of diameter changes (Cheng et al., 2012), these large number of grown particles might through the subsequent coagulation process to grow into larger size range (Seinfeld and Pandis, 2006), where they could contribute more efficiently to the particle light scattering as well as mass concentration. For example, at 16:00 LT, significantly increase in scattering size distribution within nearly the full size range from about 40-900 nm can be observed at both sites, especially at SDZ."

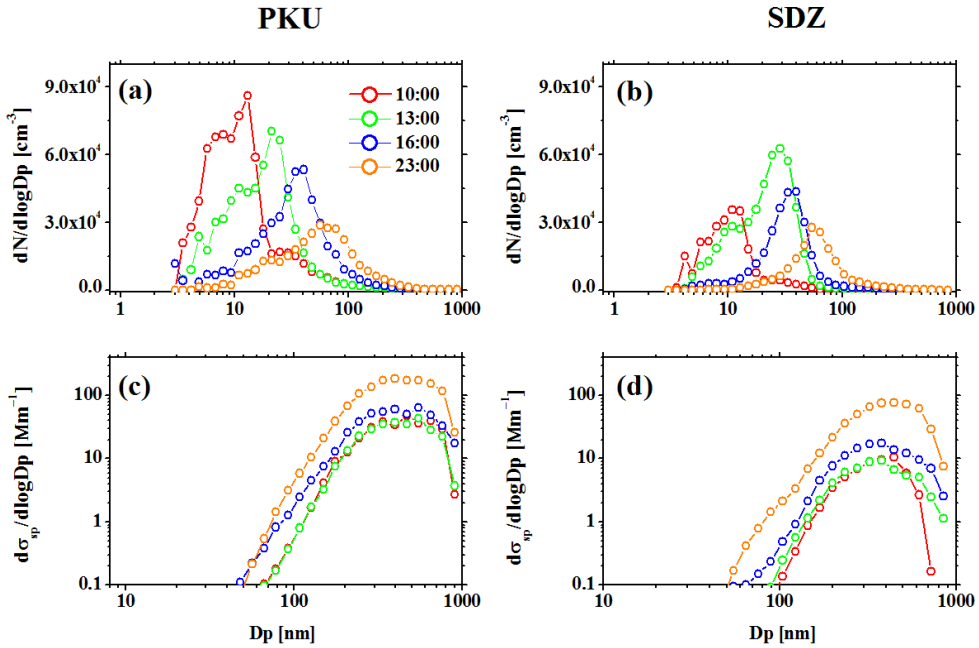

Figure 5. Evolutions of (**a&b**) the particle number size distributions measured by TDMPS and (**c&d**) the simulated particle light scattering size distributions at 550 nm from about 10:00 (local time) to midnight (23:00) on September 26, 2008. Left and right panels represent the PKU and SDZ sites, respectively.

Cheng, Y. F., Su, H., Rose, D., Gunthe, S. S., Berghof, M., Wehner, B., Achtert, P., Nowak, A., Takegawa, N., Kondo, Y., Shiraiwa, M., Gong, Y. G., Shao, M., Hu, M., Zhu, T., Zhang, Y. H., Carmichael, G. R., Wiedensohler, A., Andreae, M. O., and Pöschl, U.: Size-resolved measurement of the mixing state of soot in the megacity Beijing, China: diurnal cycle, aging and parameterization, Atmos Chem Phys, 12, 4477-4491, 10.5194/acp-12-4477-2012, 2012.

Seinfeld, J. H., and Pandis, S. N.: Atmospheric Chemistry and Physics. From Air Pollution to Climate Change, Wiley, New York, 2006.