Investigation of new particle formation at the summit of Mt. Tai, China

Ganglin Lv et al.

We are very grateful to the detailed comments offered by the Referee #2. We have revised the manuscript accordingly, and listed below in red are our point-by-point responses.

Response to Referee #2

1. The language of this manuscript is far from a scientific publication. It is strong recommended that the manuscript should be carefully revised, probably by a native English speaker. There are many grammatical mistakes, incorrect omissions and collocations, even in the abstract, including but not limited to the following items.

Answer: Thanks. The manuscript has been revised by the native English speaker. The sentences have been marked in red.

(1) Page 1, line 17, "appeared" should be "appears"

Answer: We have corrected the word. However, as we revised the abstract, the part which includes this word has been removed.

(2) Page 2, line 3, there is a "that" missing after "exhibited".

Answer: We have corrected it.

(3) Page 2, line 6, "Recent decades" should be "In recent decades".

Answer: Thanks, we have corrected it.

(4) Page 2, line 17, should "after then" be "after that"?

Answer: We used "thereafter" to replace "after then".

(5) Page 3, line 10, does the author want to say "What's the contribution of estimated gaseous sulfuric acid on nucleation and growth processes?"

Answer: Yes, we appreciated the better expression you offered and we have corrected this sentence. However, the discussion for contribution of sulfuric acid has been removed in the revised manuscript, so this sentence has also been removed. Thanks very much.

(6) Page 5, line 17, "in Table 3 it also exhibited the characteristics comparison between Mt. Tai and some other typical recent researches in China." should be "In table 3, it is also exhibited that the characteristics…"

Answer: We have corrected the sentence.

(7) Page 6, line 1. "the sampling season".

Answer: We have revised the word as the "the observation period". However, the related sentence has been removed in the revised manuscript.

(8) Page 6, line 15. “Although gaseous sulfuric acid formed through sulfur dioxide photochemical reactions was involved in NPF, neither high sulfur dioxide being strong NPF nor low value limiting NPF burst.” This sentence is too vague to be understood.

Answer: We wanted to show that the sulfur dioxide concentration didn’t directly affect the occurrence of NPF, although sulfuric acid could be formed through the photochemical reactions of sulfur dioxide. In the revised manuscript, the related sentence has been removed.

(9) Page 7, line 18. “Figure 3 picked 40 days continuous data from...” can be revised as “Figure 3 exhibits the continuous data from…”.

Answer: We appreciated the better expression you made. However, the related sentence has been removed in the revised manuscript.

(10) Page 7, line 21, “exhibited” should be “exhibits”. “This result was in accordance with…” should be “This result is in accordance with…”

Answer: Thanks, and we have corrected them. However, the related part has been removed in the revised manuscript.

(11) Page 7, line 23, better to substitute “improve” with “increase”.

Answer: Thanks, and we have corrected it.

(12) line 29, “Existent of ozone could quantify the oxidation capacity and photochemical activities in the atmosphere, directly reacting with related species such as VOCs and indirectly affecting sulfuric acid formation via hydroxyl radical production”. Incorrect adverbial use.

Answer: We have revised this sentence as “O_3 has been considered to quantify the oxidation capacity and photochemical activities in the atmosphere, directly reacting with related species such as VOCs and indirectly affecting sulfuric acid formation via hydroxyl and hydroperoxy radicals” in the revised manuscript.

(13) Page 8, line 1. Should be “it is found that…”. “revealed” should be “experienced”.

Answer: We have corrected them. However, the related sentence has been removed in the revised manuscript.

(14) Page 8, line 3. What does “made for” mean here?

Answer: The “made for” means “be in favor of”. However, we have revised the sentence as “favorable meteorological conditions could promote the occurrence of the NPF when precursors were insufficient in the atmosphere” in the revised manuscript.

(15) Page 8, line 6. “Day-to-day analysis revealed that” should be “reveal”

Answer: Thanks, and we have corrected it. However, the related sentence has been removed in the revised manuscript.

(16) Page 8, line 10. “it meant” can be revised as “This suggests’

Answer: Yes, we have corrected it.

(17) line 12, “Particle number concentrations depending on wind direction in each mode were not obvious, and none of directions always showed significantly higher or smaller particle concentrations and it had clear difference with reports in Nanjing.” This sentence is too vague to be understood.
Answer: New discussion is in the revised manuscript, please see in page 10, line 22-26. In the revised manuscript, we found that the accumulation mode particles showed a high level of concentration when the corresponding wind was in the northeast, which could partly explain the less occurrence of NPF from the northeast wind direction. In addition, we found that the nucleation and Aitken modes particles were approximate possibility in each wind direction if there was not the NPF event, suggesting that few direct particle sources for nucleation and Aitken modes particles existed around the observation site.

(18) line 14, should be “this suggests that few direct…”
Answer: We appreciated the better expression you made. But we have rewritten the related sentences, please see in page 10, line 22-26.

(19) line 18, “illustrated” should be “illustrates”.
Answer: Thanks, and we have corrected it.

(20) Page 9, line 2. Revised as “Fig. 6 illustrates the data from…”
Answer: We have corrected it. However, the related sentence has been removed in the revised manuscript.

(21) Page 10, line 5. “In contrast” instead of “by contrast”.
Answer: We have corrected it. However, the related sentence has been removed in the revised manuscript.

(22) line 11, what does “background total nucleation particles” mean?
Answer: The “background total nucleation particles” means “the particle number concentration of 3-20 nm without the effect of NPF events or other air mass transport”. In the revised manuscript, the related sentence has been removed.

(23) line 30. “It revealed that…” What does “it” here stand for?
Answer: “it” stands for “the day on 11 November 2014” in the paper. However, the related discussion has been removed in the revised manuscript.

2. Page 1, line 25, “$PM_{2.5}$ variation was always in accordance with particle total volume concentration.” This is nearly common sense. I don’t understand why this can be a conclusion in this paper.
Answer: We have deleted it in the revised manuscript.

3. Page 3, line 10. I am not sure whether the third point the author made here can be a real “scientific question”. The author may need to contribute some in-deep thoughts here.
Answer: Thanks. The discussion related to particle behaviors has been removed in the revised manuscript.

4. Page 4. The authors need to define all the parameters used in the equations.

Answer: We have made the modification in the revised manuscript.

5. Page 4, line 15. Why the F_{growth} can be neglected in this study? Does the author have evidence on this?

Answer: We have added a brief explanation in page 4, line 25-26. Based on particle size distribution data and duration of nucleation process, we first estimated the range of growth rate and found that particles rarely grow beyond 20 nm before formation ended. In our study, most newly formed particles grew slowly during the NPF events. In some cases of NPF events, particles grow faster but its duration was fairly short. As a result, the F_{growth} could be neglected in this paper. In addition, many reported papers such as Dal Maso (2005), have also neglected the growth loss.

6. Page 4, line 23. The paper written by Mikkonen et al. has provided a more precise H2SO4 estimation equation, in which another two parameters, CS and RH were used. Why don't the author use this one?

Answer: Thanks very much. In our revised manuscript, we have used the non-liner type proxies for sulfuric acid in Sect. 2.3.2, and both CS and RH have been taken into consideration.

7. Page 6, line 10. "NPF events could be observed in each month, and frequent NPF occurrence was in campaign ĐŸ which showed the frequency of 56 % (others were only 21 % by contrast). It could be interpreted that campaign Đ ˛ E and Đ1 were in rainy and foggy season, and such wet condition seemed adverse to NPF." The campaign Đ ˛ E, ĐŸ and Đ´l should be defined before.

Answer: We have added them in the introduction (page 3, line 11) and discussion (page 5, line 21-22).

8. Page 5, line 24. "Another notable period was from 10 October to 18 October 2014, during which it had frequent NPF events and most of formation rates were larger than 75th percentile (20.61 cm-3 s-1). It could be associated with specific atmospheric conditions because of sudden temperature drop." Why the sudden temperature drop increased the frequency of NPF?

Answer: Sudden temperature drop might increase the coal or biomass burning in the region, which possibly increased the emission for precursors. The elevated precursor concentration was in favor to the occurrence of NPF. However, we consider that this phenomenon is lack of accurate evidence, so the related discussion has been removed in the revised manuscript.

9. Page 6, line 1. "Reasons for our large value were possibly not only related to geographically wide mountaintop location, but also sampling season impacts and size range difference for calculating formation rate." What does the "geographically wide mountaintop location" mean? What specifically are the "season impacts" and "size range difference"?

Answer: The "geographically wide mountaintop location" means "there is not the obvious obstacle around the mountain-top observation site". The wide landform may be in favor of precursor transport from the other locations. The observation site in our study is the summit of Mt. Tai without obstacles nearby, where air mass transport may be intensive. The "geographically wide mountaintop location" may not be an appropriate use, and the new discussion is in page 6, line 17-19.

The "season impacts" means "influence of the different season", and the "size range difference" means "the different particle interval for calculating the formation rate, such as the J_3 and J_5". Because they may not be the special characteristics of Mt. Tai, we have removed the results in the revised manuscript.

10. Page 6, line 20. Why it didn't exhibit significant distinction in the condensation sink values between NPF and non-NPF days? The author needs to provide a clear explanation.
Answer: We have added a more detailed analysis of CS on NPF days and non-NPF days in Set. 3.2.1, please see in page 7, line 12-23 and page 8, line 6-9.

11. Page 6, line 30. The uncertainty is not only from the solar radiation data, but also from the calculation itself as all the parameters for the equations are estimated based on the data in EU, not in China.
Answer: We have made a new discussion related to the sulfuric acid in Set. 3.2.1, please see in page 7, line 24-31 and page 8, line 1-5. In the revised manuscript, we compare the estimated sulfuric acid proxy concentration with reports in China rather than in EU.

12. Page 8, line 1. "it found ozone concentration revealed slight drop during nucleation process on many NPF days and ozone consumption reactions might take place." What does the author mean here? The ozone concentration decreased because of the reactions during nucleation process? Why is that?
Answer: There may be confused expression in our manuscript, and we have made a more detailed and accurate discussion of ozone in Set. 3.2.2, seen in page 9, line 17-30. The diurnal variation of ozone concentration on NPF days showed a slight drop near the sunset. During this period, the residual ozone, with the high concentration at the summit of Mt. Tai, might involve in sulfuric acid formation via producing the hydroxyl and hydroperoxy radicals under the solar radiation condition. The decreased ozone concentration is not caused by the nucleation, but because of the production of the hydroxyl and hydroperoxy radicals or ozone photolysis.

13. Page 8, line 3. "our statistical results showed that NPF preferred to occur on clear or partial cloudy daytime." What is the statistical evidence here?
Answer: In this study, approximately 90 % of all the NPF events occurred on the clear or partial cloudy daytime, seen in page 10, line 2-3.

14. Page 8, line 6. "Day-to-day analysis revealed that temperature and relative humidity always had cyclic variation, and NPF events preferably occurred on high temperature and low relative humidity conditions. High temperature and low humidity could promote vertical transportation and photochemical reactions in the atmosphere." I don't think this is something that can be "revealed". NPF always occurs in the middle of the day when temperature is higher and humidity is lower, because it is driven by photochemistry. This phenomenon cannot be logically concluded as high temperature and low relative humidity favour the NPF.
Answer: Our previous understanding for temperature and relative humidity might be one-sided, and we have made a new detailed discussion of temperature and relative humidity in Set. 3.2.3, seen in page 10, line 3-17. In the revised manuscript, we compare the average diurnal variations of the temperature and relative humidity during NPF days and non-NPF days. The results showed that the lower temperature and relative humidity conditions seemed to be favorable for the occurrence of the NPF.

15. Page 8, line 15. "It could suggest that few direct particle pollution sources existed around observation site and nucleation might be the primary source for particles on Mt. Tai." I don't

understand how the author made this conclusion. If there is no local source nearby, particles can be from transport from other regions.

Answer: There might be confused expression in our manuscript, and we have rewritten this part in 10, line 22-26. In this study, we mainly want to discuss the particle number concentration distribution as the function of wind direction. During all the observation days, the accumulation mode particles showed the higher values when the wind came from northeast. So we speculate that this exceptional phenomenon may partly contribute to the less NPF occurrence in the northeast wind direction. In addition, we found that the nucleation and Aitken modes particle in all wind directions were almost evenly distributed if there was not NPF events, and none of directions showed significantly higher or smaller values. However, there were uneven and similar distributions between nucleation and Aitken modes particles during all the observation days. These phenomena suggest that NPF events possibly result in above difference. In addition, wind directions just reflect a local situation rather than the long-distance transport. If there were local point sources around observation sites, some directions should be obviously higher particle (nucleation and Aitken modes) concentrations. So we speculate that there are few direct local sources for nucleation and Aitken modes particles around the site.

16. Page 8, line 24. "Compared with air masses coming from cleaner western parts of China, air masses going through Beijing et al. polluted areas had more complicated components and enhanced NPF events." Is there any evidence in this paper to say so?

Answer: We have revised the related part, and the new detailed description could be seen in page 11, line 1-14. During all the NPF days, continental air masses accounted for 80 % of the total air masses, among which four-fifths passed though polluted areas (Beijing, Hebei Province, Shanxi Province, Henan Province, Shaanxi Province) before reaching the observation site. In contrast, 63 % of the total air masses were continental air masses during all the non-NPF days, among which only two-fifths passed though polluted areas before reaching the observation site. Most of the continental air masses on non-NPF days came from the south (cleaner part of China) or transported over Bohai Sea and Yellow Sea. From the statistical results, it seems plausible that air masses going through the polluted areas could increase the occurrence of NPF. When air masses passed though the polluted areas, it might bring precursors or motivating substances for NPF.

17. Page 8, line 29. "Hence NPF events with local continental backward trajectories were more vulnerable to local point sources." This is confusing. The author just discussed that there is few direct sources around the observation site. How does the author make this conclusion then?

Answer: There might be confused expression in our manuscript. The afore-mentioned conclusion was based on the discussion of particle number concentration distribution with wind directions, and there were few local point sources for nucleation and Aitken modes particles around the observation site. Air mass backward trajectories were based on the long-time air mass transport. In this study, all the local air masses during NPF days were upward, which might bring local pollutants to the sites to affect the NPF. However, we have removed this conclusion in the revised manuscript because a few local air mass back trajectories were observed in this study.

18. Page 9, line 9. Why "the higher atmospheric humidity in campaign ĐE¸ might enhance the sticking possibility of particles for molecules"? Does the author refer to liquid phase or the phase change due to water content?

Answer: Many literatures, such as Mikkonen et al (2011), reported that high RH might increase the sticking probability of molecules to existing particles. In our study, we didn't consider the liquid phase or the phase change. However, the discussion for $PM_{2.5}$ variation has been removed in the revised manuscript,

19. Page 9, line 17. "But particles recombination in close sizes could contribute to the growth after nucleation and higher PM2.5 within limiting values possibly increased this possibility." Coagulation of particles within the nucleation mode is always negligible under ambient condition. The author needs to provide strong evidences to prove that it is important in this study.
Answer: In the revised manuscript, the discussion related to $PM_{2.5}$ and particle growth has been removed.

20. Page 10, line 10. The author try to calculate the particle density on clean days here. In this case, the author needs to prove that the PM2.5 measurement was very accurate at low concentration level. Also, the particle growth factor should be considered if there is no dryer before the PM2.5 monitor.
Answer: Thanks very much. You have given me a very important instruction for calculating the particle density, and we think our instrument may not meet requirement for very accurate measurement at low concentration level and we will try to seek other method for calculating the particle density. However, the related discussion for particle behaviors under clean and polluted conditions has been removed in the revised manuscript.

21. Page 10, line 17. "NPF had small impact on total volume concentration on polluted day, which might be related to large background fine particles." I don't know what the author wants to express here.
Answer: On the polluted days with elevated $PM_{2.5}$ concentration, the fine particles may account for the major volume of the total particle volume concentration. However, the fine particle concentration changed a little before and after NPF events on the polluted days, suggesting that the NPF events made the minor contribution to the fine particles and the total particle volume concentration. In the revised manuscript, the related discussion has been removed.

22. Page 10, line 23, is there a definition of "haze day" before? If it means PM25 > 75 ug m-3, why the average was only 64 ug m-3?
Answer: In our revised manuscript, we use an accurate "hazy episodes" to replace "haze day". Because the occurrence of NPF was only observed in the daytime, so the "hazy episodes" might be fitter in our study. The hazy episode can be identified when the atmospheric visibility is less than 10 km and the RH is less than 80 % simultaneously, seen in page 11, line 17-19. The value of 64 ug m^{-3} just represented the average value at the summit of Mt. Tai. In fact, the average $PM_{2.5}$ concentration on hazy days/episodes at the foot of mountain is much larger than 64 ug m^{-3}.

23. Page 11, line 8. "Reason for the former was possibly clusters recombination, and the latter decrease might be on account of energy threshold (i.e., nucleation barrier) and atmospheric scavenging." Again, the author need to provide very strong prove that the coagulation of clusters is very important in this study.
Answer: Thanks. In the revised manuscript, the discussion related to particle behaviors has been

removed.