

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

Ganglin Lv et al.

5 We are very grateful to the detailed comments offered by the Referee #1 and Referee #2. We have revised the manuscript accordingly, and listed below in red are our point-by-point responses.

## Response to Referee #1

10 1. The language requires polished. I highly suggest that the authors ask a native speaker to edit this manuscript. Some sentences in this manuscript are too obscure to understand.

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

2. P1, Line 14, campaign I and II seem overlapped. Please check the campaign period.

15 Answer: We have checked the campaign period, and there was a spelling mistake in campaign I. The right period of campaign I was from 25 July to 24 August 2014, and we have corrected it.

3. P1, Line 18, Mt. Tai CANNOT show larger formation rate. Besides “larger formation rate” is misleading. this work only studied less than one year NPF, it’s difficult to conclude the formation rate here is large. Considering the gas concentration, the FR for clean site should be lower.

20 Answer: Thanks. We have rewritten the related discussion in page 6, line 9-19 in the revised manuscript. The observed NPF events at the summit of Mt. Tai were indeed very strong in our study, with the  $J_3$  of  $7.10 \pm 5.39 \text{ cm}^{-3} \text{ s}^{-1}$ . The deep-analysis for its mechanisms might need the precise measurement for precursors, and we couldn’t make the accurate explanation for it because of limited measurement instruments. However, intensive precursor transport in region (eastern China) and enhanced photochemical activities at the summit of Mt. Tai could, at least partly, explain the higher formation rate values.

4. P1, Line 22, what does “limited higher PM 2.5” mean here? Usually higher particle contribution inhabits the new particle formation. Anyway, I guess the authors were trying to say that during the relatively polluted days, the GR is higher? It’s because of the gas concentration, but has nothing to do with PM concentration.

30 Answer: We appreciated the better understanding you made for “limited higher PM<sub>2.5</sub>”, and we actually wanted to express the higher growth rate under the relatively polluted environment. However, the discussion of PM<sub>2.5</sub> and particle growth has been removed in the revised manuscript.

5. P1, Line 20, what does “proxy” mean?

35 Answer: In the manuscript, “proxy” means “proximity for sulfuric acid”. In our study, direct measurement for gas-phase sulfuric acid was unavailable. Instead, the proxy for sulfuric acid was estimated based on solar radiation, sulfur dioxide, condensation sink and relative humidity.

6. P1, Line 22, recombination? Do author want to say “coagulation”?

40 Answer: Thanks. We actually wanted to show the “self-coagulation”. However, the related sentences have been removed in the revised manuscript.

7. P1, Line 23, “haze” is inaccurate.

Answer: Thanks. We have replaced “haze” to “hazy”

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8. P2, Line 7, change “around” to “over”, what does “refer to” mean?

Answer: Thanks. We have corrected “around” to “over”, and we have also changed the “refer to” to “include”.

9. Session 2.1, more information about the site should be provided, e.g. the height of the site or inlet from ground.

Answer: We have added a more detailed description about the site, seen in page 3, line 17-26. The observation site is almost the peak of Mt. Tai, so the height of the site is near 1534 m ASL. All the instruments were installed inside a large container, sampling through short inlet tubes from the container at a height of about 3 m above the ground level.

10. P3 Line 25, “Its measurement range of...” should be rewritten.

Answer: Thanks. We have rewritten the related sentences in Sect. 2.2, please see page 4, line 3-5.

11. Session 2.3.2, the sulfuric acid estimation method used here already has very large uncertainty. Besides, the accuracy of the radiation from HYSPLIT model is far from enough for sulfuric acid estimation.

Answer: In our revised manuscript, we have used the non-linear type proxies for sulfuric acid estimation in page 5, line 8-14, and both CS and RH have been taken into consideration in the new estimation method. In this study, direct measurement for solar radiation was not possessed, and the meteorological station in China did not have the related data as well. There were not studies which reported the approximate values at the adjacent locations. As the result, the HYSPLIT model is the only available for solar radiation at the summit of Mt. Tai. We made the comparisons between the reported solar radiation and estimated solar radiation from HYSPLIT in some other locations. Results showed the absolute value of solar radiation from HYSPLIT might involve some error with the real solar radiation in the atmosphere, but its diurnal variation pattern could be believable to a certain extent. Except for the calculated sulfuric acid proxy concentrations in page 7, line 24-31 and Table 1, most of results in the revised manuscript were based on the comparison between NPF days and non-NPF days. The comparison method could weaken the effect of absolute solar radiation on the results. In addition, although the calculated sulfuric acid proxy concentrations have some error, its magnitude ( $\sim 10^6$  orders of magnitude for initial sulfuric acid concentrations at the summit of Mt. Tai,  $\text{cm}^{-3}$ ) should be believable.

12. Equation 6, the literatures here are old. For the current knowledge, sulfuric acid is considered to contribute to the nucleation, but negligibly to the particle growth. The authors should rethink about the discussion about this.

Answer: Many recent researches, such as Meng et al. (2015), showed that sulfuric acid made a minor contribution on particle growth and the oxidation production of VOCs accounted for the major contribution. In consideration of the inaccurate estimation of absolute sulfuric acid values, the discussion of sulfuric acid contribution on the NPF has been removed in the revised manuscript.

13. P5, Line 10, change “each” to “every”

Answer: We have corrected it.

14. Session 3.1, authors should analyze what controls the occurrence of NPF, source or sink?

Answer: Thanks very much. Multiple factors affecting the occurrence of NPF have been added in Sect. 3.2, page 6-11, including condensation sink, sulfuric acid proxy, sulfur dioxide, ozone, temperature, relative humidity, wind direction and air mass transport. In Sect. 3.2.1, we focus on the effects of sulfuric acid source and condensation sink, seen in page 7, line 1-31 and page 8, line 1-15.

15. Fig. 1, y should be  $\text{SO}_2 \cdot \text{OH}$  to present NPF source. Comparing  $\text{SO}_2$  with CS makes no sense.

Answer: Thanks.  $\text{SO}_2 \cdot \text{OH}$  may not be possessed in this study, but we use the sulfur dioxide proxy concentration as the Y axis in Fig. 2 to compare the source and sink on NPF days and non-NPF days.

16. Fig. 3, change the color bar so that one can see the “banana curve” clearly.

Answer: We have changed the color bar and used the ORIGIN 9.0 to make the contour plot in our revised manuscript.

17. Session 3.2, because the sulfuric acid estimation has large uncertain, author should reconsider how to analyze this part.

Answer: Thanks. As the 11<sup>th</sup> answer, we focus on the comparisons between NPF days and non-days in the revised manuscript, and avoid involving absolute values as possible as we can. Detailed discussion was seen in Sect. 3.2.1, page 7, line 24-31 and page 8, line 1-30.

5 18. P7, Line 22, this interpretation has no evidence. It's more from author's guess, not from data. There are a few interpretations like this, e.g. line 25.

10 Answer: Thanks. There might be confused expression in our manuscript, and new discussion for sulfur dioxide was seen in page 9, line 1-16. Because photochemical reactions of SO<sub>2</sub> are the major source for sulfuric acid at the summit of Mt. Tai (direct sulfuric acid could be neglected) and NPF was indeed favorable to the high SO<sub>2</sub> concentration, it is plausible that higher SO<sub>2</sub> concentration can increase the possibility of rich precursors for NPF.

19. Session 3.4, PM<sub>2.5</sub> is not directly related to NPF. Again, it should be discussed that it is source or sink that control the occurrence of NPF.

15 Answer: In the revised manuscript, the discussion for PM<sub>2.5</sub> variation has been removed. Instead, we focused on the NPF event during hazy episodes, and one NPF event on 11 November 2014 was analyzed in detail to explore the factors affecting its occurrence. Please see in Sect. 3.3, page 11, line 15-31 and page 12, line 1-18.

20. Session 3.5, how to combine the NAIS and WPS data. Especially these two instruments have overlapped size range. Maybe this should be included in the experimental session.

20 Answer: Thanks very much. Combination of the NAIS and WPS is a very good suggestion. It was a pity that the particle number size distributions measured by two instruments matched not very well in this study, and the WPS data were discontinuous because of frequent instrument maintaining. Therefore, we mainly used the NAIS data in the analysis of the NPF events, and the WPS data were the assist. However, we can adopt the combination of the NAIS and WPS in our future study.

25 21. Session 3.6, the definition of "haze" here is unclear. Visibility is not a crucial criterion for haze. The PM concentrations are similar for so called haze and non-haze days. Can it be haze or fog? Also it seems redundant and reduplicated to session 3.5.

30 Answer: In the revised manuscript, 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 fog episode is identified when the atmospheric visibility is less than 10 km and the RH is more than 90 %. The crucial criterion between haze and fog is the RH. In the revised manuscript, we focus on the factors affecting the occurrence of NPF, and the related discussion for particle behaviors has been removed.

### 35 References

Meng, H., Zhu, Y., Evans, G. J., Jeong, C. H., and Yao, X.: Roles of SO<sub>2</sub> oxidation in new particle formation events, J. Environ. Sci. (China), 30, 90-101, doi:10.1016/j.jes.2014.12.002, 2015.

## Response to Referee #2

1. The language of this manuscript is far from a scientific publication. It is strongly 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: Thanks, 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: Thanks, 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.

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(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<sub>3</sub> 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.

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

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

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(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.

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(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.

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(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.

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(18) line 14, should be “this suggests that few direct...”

Answer: We appreciated the better expression you made. However, we have rewritten the related sentences, please see in page 10, line 22-26.

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(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.

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(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.

5 (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.

10 2. Page 1, line 25, “PM<sub>2.5</sub> 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: Thanks, we have deleted it in the revised manuscript.

15 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. Please see page 4, line 20-29 and page 5, line 1-14.

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

Answer: Yes. 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  $F_{\text{growth}}$  could be neglected in this paper. In addition, many reported papers such as Dal Maso (2005), have also neglected the growth loss.

30 6. Page 4, line 23. The paper written by Mikkonen et al. has provided a more precise H<sub>2</sub>SO<sub>4</sub> 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-linear type proxies for sulfuric acid in Sect. 2.3.2, and both CS and RH have been taken into consideration.

35 7. Page 6, line 10. “NPF events could be observed in each month, and frequent NPF occurrence was in campaign D<sup>Y</sup> which showed the frequency of 56 % (others were only 21 % by contrast). It could be interpreted that campaign D<sub>1</sub> E and D<sub>1</sub> were in rainy and foggy season, and such wet condition seemed adverse to NPF.” The campaign D<sub>1</sub> E, D<sup>Y</sup> and D<sub>1</sub> should be defined before.

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

40 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<sup>-3</sup> s<sup>-1</sup>). 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.

45 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: Thanks. 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: Thanks. 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: Yes. 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: Thanks. 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 particles) 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: Thanks. 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 through polluted areas (Beijing, Hebei Province, Shanxi Province, Henan Province, and 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 through 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 through 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: Thanks. 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  $\Delta E_c$  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. In the revised manuscript, the discussion for  $PM_{2.5}$  variation has been removed.

19. Page 9, line 17. “But particles recombination in close sizes could contribute to the growth after nucleation and higher  $PM_{2.5}$  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: Thanks. 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 PM<sub>2.5</sub> measurement was very accurate at low concentration level. Also, the particle growth factor should be considered if there is no dryer before the PM<sub>2.5</sub> monitor.

5 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.

10 Answer: On the polluted days with elevated PM<sub>2.5</sub> 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. However, the related discussion has been removed in the revised manuscript.

15 22. Page 10, line 23, is there a definition of "hazy day" before? If it means PM<sub>2.5</sub> > 75 ug m<sup>-3</sup>, why the average was only 64 ug m<sup>-3</sup>?

20 Answer: In our revised manuscript, we use an accurate "hazy episodes" to replace "hazy 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<sup>-3</sup> just represented the average value at the summit of Mt. Tai. In fact, the PM<sub>2.5</sub> concentration on hazy days/episodes at the foot of mountain is much larger than 64 ug m<sup>-3</sup>.

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.

25 Answer: Thanks. In the revised manuscript, the discussion related to particle behaviors has been removed.

#### References

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35 Mikkonen, S., Romakkaniemi, S., Smith, J. N., Korhonen, H., Petäjä T., Plass-Duelmer, C., Boy, M., McMurry, P. H., Lehtinen, K. E. J., Joutsensaari, J., Hamed, A., Mauldin Iii, R. L., Birmili, W., Spindler, G., Arnold, F., Kulmala, M., and Laaksonen, A.: A statistical proxy for sulphuric acid concentration, *Atmos. Chem. Phys.*, 11, 11319-11334, doi:10.5194/acp-11-11319-2011, 2011.

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

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**Abstract.** To date few field observations of new particle formation (NPF) have been carried out at the high-elevation mountain sites in China. Simultaneous measurements of particle size distributions, gas species, meteorological conditions, mass concentration and chemical composition of PM<sub>2.5</sub> were performed at the summit of Mt. Tai (1534 m ASL) from 25 July to 24 August 2014 (I), 21 September to 9 December 2014 (II), and 16 June to 7 August 2015 (III), to investigate the NPF characteristics related to the relatively clean mountain-top environment. The NPF events were identified based on particle size distributions, and 66 such events were observed in a period of 164 days - corresponding to an occurrence frequency of 40 %. Formation rates of 3 nm ( $J_3$ ) and growth rates (GR) were in the range of 0.82-25.04 cm<sup>-3</sup> s<sup>-1</sup> and 0.58-7.76 nm h<sup>-1</sup>, respectively. On the average, the condensation sink (CS), O<sub>3</sub> concentration, temperature and relative humidity (RH) were lower, whereas the SO<sub>2</sub> concentration was higher on NPF days than on non-NPF days. The onset of NPF events at the summit of Mt. Tai might not be limited by the initial sulfuric acid concentration because of the approximate sulfuric acid proxy concentrations in the early morning on NPF days and non-NPF days. The analysis of back trajectory indicated that the majority of transport pathways on NPF days came from the northwest, and the air masses going through the polluted areas could increase the occurrence of NPF. Four NPF events, with the potential elevated PM<sub>2.5</sub> and trace gases concentrations, were observed during hazy episodes. As a case study on 11 November 2014, the NPF event might be driven by enhanced solar radiation at noon and the changing air mass transport with the rich precursor concentrations.

**Keywords.** New particle formation; Mountain observation; Favourable conditions; Hazy episodes

## 1 Introduction

Atmospheric aerosols play a critical role in affecting global radiation balance and climate, directly through scattering and absorption of solar radiation, and indirectly by modifying cloud properties as potential cloud condensation nuclei (CCN) (Kuang et al., 2010). Aerosol particles are involved in several atmospheric chemistry processes such as enhancing haze and

decreasing visibility, and they can also harm human health by inhalation (Han, 2012). Previous studies have showed that the nucleation of atmospheric gas-phase precursors and the subsequent growth to larger particles, widely known as new particle formation (NPF), is the largest source of atmospheric aerosol particles (Zhang et al., 2012). Field observations have exhibited that NPF typically increase the particle number concentration by a factor between two and ten (Gong et al., 2010).  
5 Model studies also revealed that NPF accounted for 5-50 % of CCN in the lower boundary layer (Spracklen et al., 2008). An in-depth study of the process of NPF and its effects could help control atmospheric aerosol pollution in China.

With the development of instruments that measure particle size distribution, NPF events have been widely observed all over the world in recent decades. These observation sites included northern-most sub-arctic, remote boreal forests, industrialized agricultural regions, high-iodine coastal environments, and polluted urban areas (Dal Maso et al., 2002). The  
10 frequency of NPF events varies significantly between locations. Hallar et al. (2011) reported that NPF events in urban areas such as Pittsburgh occurred on about 35-50 % of all days, while remote background sites in Finland and Sweden only showed such events on 2-27 % of days. Manninen et al. (2010) found that the frequency of NPF days ranged from 21 % to 57 % based on the observations at twelve field sites around Europe, and the number of observed NPF days was closely related to the regional atmospheric conditions.

In the past decade, many campaigns and studies on NPF have been carried out in China. In 2004 Wiedensohler et al. (2004) first reported the observation of NPF events in China using a Twin Differential Mobility Particle Sizer (TDMPS). Soon after, Liu et al. (2008) made observations of NPF at a rural/coastal site in XinKen (Guangdong Province). In 2005, Gao et al. (2009) investigated the occurrence of NPF in a suburban environment in the Yangtze River delta using a Wide-range Particle Spectrometer (WPS). Thereafter, several observations of the NPF have been reported in urban/suburban/rural environments  
20 around China (An et al., 2015; Wang et al., 2011). However, there have not been many observations of NPF on mountain-top sites in China so far. Zhang et al. (2016) reported the occurrence of NPF on Mt. Huang (1840 m ASL) with the WPS instrument from April to July 2008. However, these results had significant limitation in measurement methods and seasonal variation. It is clear that multi-season observations using nanometer-scale instruments would be essential and valuable for the NPF research in mountain environments.

Previous researches have shown that sulfuric acid, ammonia, organic vapors, and iodide species in the atmosphere were involved in nucleation process in specific conditions. Gaseous sulfuric acid was the most critical candidate that participated in binary, ternary and ion induced nucleation (Boy et al., 2005; Wang et al., 2011; Zhang, 2010; Saunders et al., 2010; Allan et al., 2015). It has also been shown that the nucleation rate is a function of sulfuric acid concentration with a power dependency exponent, whose value of exponent varies significantly between different nucleation theories (Kulmala et al.,  
30 2006; Wang et al., 2011). However, the occurrence of NPF cannot be determined by a single sulfuric acid factor, and other factors such as pre-existing particles, trace gases, meteorological conditions, and air mass transport should also be considered. However, comprehensive investigations of these factors affecting the occurrence of NPF in China are mainly

conducted in urban/suburban/rural environments. Since the mechanism of NPF under the heavily polluted conditions has significant difference to that under the relatively clean conditions, the intensive and comprehensive investigation of NPF on mountain-top sites may be very important.

Particle formation and growth rates vary with field environments. Kulmala et al. (2004) reviewing a number of studies found that the typical formation rate was in the range of  $0.01\text{-}10\text{ cm}^{-3}\text{ s}^{-1}$ . In urban areas it may be about  $100\text{ cm}^{-3}\text{ s}^{-1}$ , while in coastal zones it can be as high as  $10^4\text{-}10^5\text{ cm}^{-3}\text{ s}^{-1}$ . Typical growth rate of newly formed particles ranges from 1 to  $20\text{ nm h}^{-1}$ , and at some coastal areas it is as high as  $200\text{ nm h}^{-1}$ .

In this paper, we present the results of the intensive field campaigns at the summit of Mt. Tai (1534 m ASL) related to the relatively clean mountain-top environment. This study was based on simultaneous measurements of particle size distributions, meteorological conditions, gaseous species, mass concentration and chemical composition of  $\text{PM}_{2.5}$  during three campaigns (25 July to 24 August 2014, I; 21 September to 9 December 2014, II; 16 June to 7 August 2015, III). The general characteristics of the NPF events were calculated on the basis of particle size distributions, and factors affecting NPF occurrence were discussed by analyzing condensation sink (CS), sulfuric acid, trace gas concentrations and meteorological parameters. In addition, one typical NPF event was specifically investigated to further explore the unusual NPF occurrence during hazy episodes.

## 2 Methodology

### 2.1 Site description

The observation was conducted at the summit of Mt. Tai ( $36.25^\circ\text{N}$ ,  $117.1^\circ\text{E}$ , 1534 m ASL), located in the middle part of Shandong Province, eastern China. Mt. Tai is one of the highest mountains near the East China Sea on the transport path of the Asian continental outflow (Li et al., 2011), adjacent to the Bohai Sea (B-S) and Yellow Sea (Y-S). The field site is nearly at the summit of Mt. Tai, and its surroundings are dominated by dense vegetation and mountains with few anthropogenic sources. The nearest small city, Tai'an (population: 670,000), is located about 15 km away to the south and southeast. The city of Ji'nan (population: 2,800,000), capital of Shandong Province, is 60 km to the north. During daytime, elevation of Mt. Tai reaches close to the top of the planetary boundary layer (PBL) and the site is therefore representative of the region (Zhang et al., 2014; Sun et al., 2016). All the instruments were installed inside a large container, sampling through short inlet tubes from the container at a height of about 3 m above the ground level.

### 2.2 Measurement techniques

Two types of size distribution instruments, namely neutral cluster and air ion spectrometer (NAIS) and wide-range particle spectrometer (WPS), two gas monitors ( $\text{SO}_2$  and  $\text{O}_3$ ), an instrument for mass concentration of  $\text{PM}_{2.5}$  and a monitor for chemical composition in  $\text{PM}_{2.5}$  were used in this study. In addition, meteorological parameters including air temperature (T),

relative humidity (RH), wind speed (WS), wind direction (WD) and visibility were also recorded in real time.

The NAIS is a multichannel nanometer aerosol instrument which can measure the distribution of aerosol particles and ions (charged particles and cluster ions) of both polarities simultaneously. The aerosol particle distribution of NAIS is in the size range of 2-40 nm, and the ion distribution is in the electric mobility range of 0.0013-3.2 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup> (equivalent to particle Millikan diameters of 0.8-40 nm). The instrument consists of two multichannel electrical mobility analyzer columns, one for each polarity. The aerosols are classified according to mobility and measured with an array of twenty-one electrometers per column. In this study, the total time of each measurement cycle was set at 5 min, comparing sampling intervals as follows: particles 120 s, ions 120 s and offset 60 s.

The WPS is a high-resolution aerosol spectrometer which combines a differential mobility analyzer (DMA), a condensation particle counter (CPC) and a laser light scattering (LPS). The diameter range of the WPS was from 10 to 10,000 nm, and 48 channels were used in the DMA and 24 channels were used for the LPS. The one scan time for the entire size range was set to 5 min.

The concentration of SO<sub>2</sub> in the atmosphere was measured using a pulsed ultraviolet fluorescence analyzer (Model 43C, Thermo Electron Corporation-TEC), and O<sub>3</sub> was measured using an ultraviolet photometric analyzer (Model 49C, TEC). Mass concentration of PM<sub>2.5</sub> was detected by a monitor utilizing a combination of beta attenuation and light scattering technology (Model 5030 SHARP Monitor, Thermo Fisher Scientific), and chemical composition in PM<sub>2.5</sub> was measured by Monitor for Aerosols and Gases (MARGA, ADI20801, Applikon-ECN, Netherlands). Besides, meteorological data were obtained from an automatic meteorological station (MILOS520, Vaisala, Finland).

## 2.3 Data analysis

### 2.3.1 Formation rate, growth rate and condensation sink

In this study, particles in the size range of 3-20 nm was regarded as nucleation particles, and the formation rate of nucleation mode particles,  $J_{3-20}$ , can be expressed (Dal Maso et al., 2005) as:

$$J_{3-20} = \frac{dN_{3-20}}{dt} + F_{\text{CoagS}} + F_{\text{growth}} \quad (1)$$

where  $dN_{3-20}/dt$  is the net rate of increased nucleation mode particles,  $F_{\text{CoagS}}$  is the coagulation loss and  $F_{\text{growth}}$  is the loss of particles growing out of size range. In our observation, the  $F_{\text{growth}}$  term could be neglected because particles growing beyond 20 nm before formation ended was relatively rare. In addition, the formation rate of 3 nm particles,  $J_3$ , was also calculated from the NAIS data (Sihto et al., 2006; Kulmala et al., 2012) using the equation:

$$J_3 = \frac{dN_{3-6}}{dt} + \text{CoagS}_{Dp=4 \text{ nm}} \cdot N_{3-6} + \frac{1}{3 \text{ nm}} \text{GR}_{3-6} \cdot N_{3-6} \quad (2)$$

Here  $\text{CoagS}_{Dp=4 \text{ nm}}$  represents the coagulation sink of 4 nm particles, an approximation for the interval of 3-6 nm.  $\text{GR}_{3-6}$  and  $N_{3-6}$  denote the growth rate and particle number concentration between 3 and 6 nm, respectively.

The particle growth rate, GR, was determined by the maximum concentration method (Kulmala et al., 2012), that is:

$$GR = \frac{\Delta D_m}{\Delta t} = \frac{D_{m2} - D_{m1}}{t_2 - t_1} \quad (3)$$

where  $D_{m1}$  and  $D_{m2}$  are the geometric median diameter of representative particles at the start time  $t_1$  and the end time  $t_2$ , respectively.

5 Condensation sink, CS, determines the rate of molecules condensing on the pre-existing aerosols, and it is given by (Dal Maso et al., 2005; Kulmala et al., 2001):

$$CS = 2\pi D \sum_i \beta_{Mi} D p_i N_i \quad (4)$$

where  $D$  is the diffusion coefficient for sulfuric acid, and  $\beta_M$  is the size-dependent transitional correction factor.

### 2.3.2 Sulfuric acid proxy

Direct measurement of gas-phase sulfuric acid wasn't available in this study. Instead, the predictive proxy for sulfuric acid ( $[H_2SO_4]$ ) could be estimated based on solar radiation,  $SO_2$  concentration, CS and RH (Mikkonen et al., 2011):

$$10 [H_2SO_4] = 8.21 \cdot 10^{-3} \cdot k \cdot SR \cdot [SO_2]^{0.62} \cdot (CS \cdot RH)^{-0.13} \quad (5)$$

Here  $k$  is temperature-dependent reaction rate constant, and solar radiation is estimated from Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPPLIT) Model in Air Resources Laboratory. The absolute values of solar radiation from HYSPPLIT may involve some error with the real solar radiation in the atmosphere, but its diurnal variation pattern is believable.

## 15 3 Results and discussion

### 3.1 Classification and characteristics of the NPF events

Basically, a NPF event could be defined as a distinct burst of new nucleation mode particles and subsequent growth of particles to larger size over a period of time (Dal Maso et al., 2005; Hallar et al., 2011; Wang et al., 2014a; Xiao et al., 2015). For the Mt. Tai observation in this study, neutral particles generally accounted for more than 95 % of the total particles during NPF events, so a detailed discussion of ions will not be included in this paper.

In this study, three campaigns (25 July to 24 August 2014, I; 21 September to 9 December 2014, II; 16 June to 7 August 2015, III) were conducted at the summit of Mt. Tai. For the valid observations of 164 days, the NPF events occurred on 66 days, corresponding to an occurrence frequency of 40 %. NPF events were observed in every month, and the frequent occurrence of NPF was in the campaign II which showed the frequency of 56 % (the others were only 21 % by contrast). The different results could be attributed to the rainy/foggy phenomena in the campaign I and III which hindered the NPF. In this study, we defined the observed start time of the NPF events based on the significant enhancement of the  $N_{3-6}$ . As the result, approximately 95 % of the NPF events were initiated at 8:00-11:00 LT (local time) at the summit of Mt. Tai, in agreement

with the previous reports in China (Guo et al., 2012; An et al., 2015; Kulmala et al., 2016; Hao et al., 2015). Previous researches explained that this time period corresponded to intensive photochemical activities, leading to potential precursors for the NPF (Hallar et al., 2011; Guo et al., 2012). Table 1 lists the calculated parameters of all the NPF events observed at the summit of Mt. Tai, such as formation rate of nucleation mode particles, formation rate of 3 nm particles, growth rate, condensation sink, mean sulfuric acid proxy concentration in the early morning (generally 6:00-9:00 LT on Mt. Tai), SO<sub>2</sub> concentration (6:00-13:00 LT), and O<sub>3</sub> concentration (6:00-13:00 LT). Table 2 summarizes the mean, median, 25th percentile, 75th percentile, minimum and maximum of these parameters on the basis of the Table 1. In Table 3, it is also exhibited the characteristics comparison between Mt. Tai and some other typical recent researches in China.

The net increase rates and formation rates of nucleation mode particles on Mt. Tai were in the range of 0.96-48.52 cm<sup>-3</sup> s<sup>-1</sup> and 1.10-57.43 cm<sup>-3</sup> s<sup>-1</sup>, respectively. Coagulation loss averagely accounted for 24.6 % of the nucleation mode particle formation. The maximal values of the net increase rate and formation rate were in the same day, and the SO<sub>2</sub> concentration also reached the maximum of 12.9±9.6 ppb on 3 December 2014. The formation rates  $J_3$  varied from 0.82 to 25.04 cm<sup>-3</sup> s<sup>-1</sup>, and the median, 25th percentile and 75th percentile were 6.15, 3.31 and 9.41 cm<sup>-3</sup> s<sup>-1</sup>, respectively. On 3 December 2014,  $J_3$  also peaked, showing an obvious sulfuric acid-controlling NPF event. As shown in Table 3, formation rate at the summit of Mt. Tai was significantly larger than 0.97-10.2 cm<sup>-3</sup> s<sup>-1</sup> at the waist of Mt. Tai Mo Shan and 0.09-0.30 cm<sup>-3</sup> s<sup>-1</sup> on the top of Mt. Huang (Guo et al., 2012; Zhang et al., 2016), but smaller than the results in Beijing and Shanghai (Xiao et al., 2015; Wang et al., 2015). In addition, the observed formation rate on Mt. Tai was a little larger than the rural/suburban environments in Table 3 (Liu et al., 2008; Yue et al., 2013; Qi et al., 2015), which was likely related to the intensive precursor transport in region (eastern China) and enhanced photochemical activities at the summit of Mt. Tai.

Growth rates GR<sub>3-20</sub> at the summit of Mt. Tai ranged from 0.58 to 7.76 nm h<sup>-1</sup>, and the median, 25th percentile and 75th percentile were 1.55, 1.15 and 2.51 nm h<sup>-1</sup>, respectively. Growth rate on Mt. Tai was comparable with some mountain observations such as 1.5-8.4 nm h<sup>-1</sup> at Mt. Tai Mo Shan, 1.42-4.53 nm h<sup>-1</sup> on Mt. Huang, and 0.8-3.2 nm h<sup>-1</sup> on Mt. Daban (Du et al., 2015; Guo et al., 2012; Hao et al., 2015; Zhang et al., 2016). Growth rates observed at the rural/suburban/urban sites behaved larger than these mountain observations shown in Table 3 (Yue et al., 2013; Liu et al., 2008; Gao et al., 2011; Qi et al., 2015; Xiao et al., 2015), suggesting that the relatively clean mountain environments might be insufficient vapors for newly formed particle growth.

### 3.2 Factors affecting the occurrence of NPF

Favorable condition of NPF events varies in the different environments. In order to further explore the factors affecting the occurrence of NPF at the summit of Mt. Tai, multiple parameters were compared between NPF days and non-NPF days. Figure 1 shows the average diurnal variations of the condensation sink, sulfuric acid proxy, sulfur dioxide, ozone, temperature, and relative humidity during NPF days and non-NPF days over all the campaigns.

### 3.2.1 Effects of condensation sink and sulfuric acid on the NPF

In principle, the observed NPF events could be considered as the result of competing between sink and source. Newly formed particles are easily scavenged by larger pre-existing particles, leading to the newly formed particles continually being reduced. On the other hand, sufficient low volatility vapors can contribute to persistent nucleation. Therefore, CS and precursor vapors are the key controlling factors associated with the NPF events. As expected, the lower CS and higher precursor concentrations would be favorable for the occurrence of NPF events, similar results shown in Wang et al. (2011) and Guo et al. (2012). In fact, one of them may be the major factor for NPF occurrence because of inter-competition between sink and source. Kanawade et al. (2014) reported that the NPF was not limited for the CS by reason of the approximate values between NPF days and non-NPF days in a semi-rural location, India. In some urban cities of China such as Qingdao, the NPF events could be observed with both of high particle loadings and precursors (Zhu et al., 2014; Kulmala et al., 2016; Xiao et al., 2015).

The hourly average CS on non-NPF days was always higher than that on NPF days (Fig. 1a), corresponding to the daily average values of  $2.0 \pm 0.5 \times 10^{-2} \text{ s}^{-1}$  and  $1.4 \pm 0.5 \times 10^{-2} \text{ s}^{-1}$ , respectively. The result indicates that the occurrence of NPF events at the summit of Mt. Tai might be limited by the lower CS condition. The general diurnal tendencies of the CS on NPF days and non-NPF days were consistent, showing the trough in the morning and the peak in the late afternoon (or at night). The hourly difference values between NPF days and non-NPF days might be decreased after 14:00 LT, such as minimal values between 18:00-20:00 LT, because of new particle formation and subsequent growth. The CS on NPF days varied from  $0.1 \times 10^{-2}$ - $28.4 \times 10^{-2} \text{ s}^{-1}$ , corresponding median, 25th percentile and 75th percentile were  $0.9 \times 10^{-2} \text{ s}^{-1}$ ,  $0.5 \times 10^{-2} \text{ s}^{-1}$  and  $1.7 \times 10^{-2} \text{ s}^{-1}$ , respectively. The result was much larger than  $0.5 \times 10^{-3}$ - $3.5 \times 10^{-3} \text{ s}^{-1}$  in Hyytiälä, Finland (Dal Maso et al., 2005), but significantly lower than many reports in China such as  $0.6 \times 10^{-2}$ - $8.4 \times 10^{-2} \text{ s}^{-1}$  in Beijing,  $0.9 \times 10^{-2}$ - $3.9 \times 10^{-2} \text{ s}^{-1}$  in Nanjing,  $0.9 \times 10^{-2}$ - $5.3 \times 10^{-2}$  in Qingdao, and  $1.0 \times 10^{-2}$ - $6.2 \times 10^{-2} \text{ s}^{-1}$  in Hong Kong (Zhang et al., 2011; Gao et al., 2012; Guo et al., 2012; An et al., 2015; Herrmann et al., 2014; Zhu et al., 2014). Overall, the environment at the summit of Mt. Tai is relatively clean with the low particle loadings.

Gas-phase sulfuric acid has been identified as an important precursor for nucleation process. Direct emission for sulfuric acid at the summit of Mt. Tai is negligible, and thus the photochemical reactions of  $\text{SO}_2$  are the significant source for sulfuric acid in the atmosphere. In this study, the mean sulfuric acid proxy concentrations (6:00-9:00 LT) of all the NPF days were in the range of  $0.52 \times 10^6$ - $25.7 \times 10^6 \text{ cm}^{-3}$ . Both curves for sulfuric acid proxy concentration peaked at noon (Fig. 1b), which was consistent with the peak time of solar radiation. The maximum hourly average sulfuric acid proxy concentration on NPF days ( $5.9 \times 10^7 \text{ cm}^{-3}$ ) was about 60 % higher than that on non-NPF days ( $3.7 \times 10^7 \text{ cm}^{-3}$ ), which indirectly reflected an intensive solar radiation on NPF days. In the early morning, the average sulfuric acid proxy concentration on NPF days was  $5.23 \times 10^6 \text{ cm}^{-3}$ , comparable with  $4.1 \times 10^6 \text{ cm}^{-3}$  in Beijing but much lower than  $2.3 \times 10^7$ - $6.4 \times 10^7 \text{ cm}^{-3}$  in Shanghai and



$6.6 \times 10^7$ - $7.8 \times 10^7 \text{cm}^{-3}$  in Nanjing (Wang et al., 2014b; Wang et al., 2015; Xiao et al., 2015). The polluted observation environments with the elevated sink might require more condensable vapors involving in nucleation, in agreement with the fact that many overseas studies observed less sulfuric acid concentration for the NPF (Dal Maso et al., 2005; Boy et al., 2005). Apart from other precursor species, lower initial sulfuric acid proxy concentration can be partly explained by the relatively lower CS at the summit of Mt. Tai.

The relationships between sulfuric acid, CS and the  $N_{3-6}$  in the early morning are shown in Fig.2. Most of the CS on non-NPF days was situated in the intermediate region ( $\sim 1.0 \times 10^{-2} \text{s}^{-1}$ ), whereas approximately 80 % of the CS on NPF days was less than  $1.0 \times 10^{-2} \text{s}^{-1}$ . Expectedly, the left region (small CS) was believed to be more favorable for nucleation than the right region (large CS). The spot distributions on sulfuric acid proxy concentration (Y axis) had no remarkable difference between NPF days and non-NPF days, which was consistent with the tendencies in Fig. 1b (6:00-9:00 LT). This phenomenon might be explained that the intensive photochemical activities frequently occurred because of the enhanced solar radiation and high oxidation capacity at the summit of Mt. Tai in the morning, which led to the sufficient vapors for nucleation burst. Therefore, we can conclude that the sulfuric acid concentration sometimes may be not the major limiting factor for the onset of NPF at the summit of Mt. Tai if the estimation for gas-phase sulfuric acid had the high confidence level.

It was noteworthy that the  $N_{3-6}$  showed a significant correlation with the sulfuric acid proxy concentration on NPF days (Fig. 2a). Lots of  $N_{3-6}$  were less than  $500 \text{cm}^{-3}$  when the sulfuric acid proxy concentration (Y axis) was lower than  $1.0 \times 10^6 \text{cm}^{-3}$ , whereas the  $N_{3-6}$  reached nearly even more than  $800 \text{cm}^{-3}$  when the sulfuric acid proxy concentration (Y axis) exceeded  $1.0 \times 10^6 \text{cm}^{-3}$ . Further investigation found that sulfuric acid proxy concentration indeed behaved a positive correlation with the  $N_{3-6}$  on many NPF days. In contrast, Fig.2b didn't exhibit similar correlation between the  $N_{3-6}$  and the sulfuric acid proxy concentration on non-NPF days, and the colors ( $N_{3-6}$ ) were almost evenly distributed on sulfuric acid proxy (Y axis).

As the vital nucleation precursor, sulfuric acid was associated with the freshly nucleation particles. As above, the positive correlation between the sulfuric acid proxy concentration and  $N_{3-6}$  at the summit of Mt. Tai was in accordance with the earlier reports (Kulmala et al., 2006; Wang et al., 2011; Guo et al., 2012). As an example on 14 October 2014 in Fig. 3, the  $N_{3-6}$  reflected a best correlation ( $R^2 = 0.975$ ) with sulfuric acid proxy concentration during NPF event (6:00-14:00 LT). After 14:00 LT, the exceptionally high  $\text{SO}_2$  concentrations (change from 2.6 ppb at 14:00 LT to 19.1 ppb at 15:00 LT) resulted in the divarication of them. In principle, the increase of sulfuric acid concentration should be earlier than the  $N_{3-6}$ . However, there were some NPF days with zero or negative time delay in this study, such as nearly zero time delay on 14 October 2014 in Fig. 3. Wang et al. (2011) explained that the fore-formed nucleation mode particles and rapid particle growth might account for such zero or negative time delay.

### 3.2.2 Effects of sulfuric dioxide and ozone on the NPF

As shown in Fig. 1c, almost all the hourly average SO<sub>2</sub> concentrations on NPF days were higher than the corresponding values on non-NPF days (except for the slightly lower values at 1:00 LT and 3:00 LT). The average SO<sub>2</sub> concentrations between 6:00-13:00 LT on NPF days and non-NPF days were 3.2 ppb and 2.6 ppb, respectively, which indicated that the NPF was indeed favorable to the high SO<sub>2</sub> concentration. Photochemical reactions of SO<sub>2</sub> are the major source for sulfuric acid at the summit of Mt. Tai, thus the higher SO<sub>2</sub> concentration can increase the possibility of rich precursors for NPF. In Fig.4, it was noteworthy that the temperature suddenly dropped from 1.3 °C to -9.4 °C on 30 November 2014. After several days, the exceptionally high SO<sub>2</sub> concentration was observed (7.1±7.2 ppb) at the summit of Mt. Tai (marked in violet block), and frequent NPF events occurred during this period. Reasons for above phenomena might be the anthropogenic pollutants (such as SO<sub>2</sub>) from the coal or biomass burning in the upwind region (Li et al., 2015a; Li et al., 2015b).

Some of the NPF events might be lack of sensitivity with SO<sub>2</sub> concentration. On 21 September 2014, for instance, non-NPF event occurred under simultaneously high SO<sub>2</sub> concentration (9.7±7.0 ppb, 6:00-13:00 LT) and CS (4.0±1.2×10<sup>-2</sup> s<sup>-1</sup>, 6:00-13:00 LT) conditions. The sink might dominate the inter-competition between sink and source on this day, which possibly limited the nucleation as the some previous reports (Song et al., 2010; Guo et al., 2012; Meng et al., 2015). Therefore, species accompanying with SO<sub>2</sub> emission, such as aerosol particles, were taken account in the investigation of NPF and SO<sub>2</sub>. Of course, the coupling of SO<sub>2</sub> and the accompanying species at the summit of Mt. Tai might be weakened a lot during air mass transport.

O<sub>3</sub> 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 (Berndt et al., 2010; Gómez Mart í et al., 2013; Sorribas et al., 2015; Guo et al., 2012). The daily average O<sub>3</sub> concentrations on NPF days and non-NPF days were 40 ppb and 47 ppb, respectively. Previous reports showed that elevated O<sub>3</sub> concentration was benefit to the occurrence of NPF (An et al., 2015; Guo et al., 2012; Zhang et al., 2016; Huang et al., 2016), but an opposite result was unexpectedly observed in the study. All the hourly average O<sub>3</sub> concentrations on NPF days were lower than that on non-NPF days in Fig. 1d. As we all know, O<sub>3</sub> is usually contributed by anthropogenic emissions on the ground. The different result in the study might be partly explained that the positive rising of O<sub>3</sub> photolysis couldn't offset the negative influence of other pollutants (accompanying with O<sub>3</sub>, such as particles). The diurnal variation of O<sub>3</sub> on NPF days showed a small trough in the early morning and the broad peak in the afternoon. The small trough was near the sunset, which might suggest that residual O<sub>3</sub> in the atmosphere was consumed and sulfuric acid was possibly formed during this period. Another remarkable feature was that the hourly difference values of O<sub>3</sub> between NPF days and non-NPF days (Fig. 1d) decreased after 10:00 LT. The decreasing difference values indirectly reflect the intensive solar radiation on NPF days because of the association between O<sub>3</sub> production and solar radiation.

### 3.2.3 Effects of meteorological conditions on the NPF

Favorable meteorological conditions can promote the occurrence of NPF when precursors are insufficient in the atmosphere (Song et al., 2010). In this study, approximately 90 % of the NPF events occurred on the clear or partial cloudy daytime, suggesting an association between solar radiation and NPF as above. The temperature of all the NPF days varied from -11.8 to 22.1 °C, and the diurnal temperature showed a cyclic variation with solar radiation. The hourly average temperature on non-NPF days ( $10.7 \pm 1.1$  °C) were always higher than that on NPF days ( $7.0 \pm 1.8$  °C) in Fig. 1e, in agreement with the studies at Mt. Tai Mo Shan and Mt. Huang, China (Guo et al., 2012; Zhang et al., 2016), and in PUY, France (Rose et al., 2015). Guo et al. (2012) indicated that the lower temperature could enhance the nucleation of sulfuric acid and water vapor. Young et al. (2007) explained that effects of lower temperature on the NPF might be related to the atmospheric vertical convection. Anyway, the occurrence of NPF at the summit of Mt. Tai seemed to favor the lower temperature condition.

The RH of all the NPF days ranged from 22 % to 95 %, and diurnal RH behaved inversely correlated with the solar radiation. The hourly average RH on NPF days ( $63 \pm 5$  %) was much lower than the corresponding value on non-NPF days ( $88 \pm 2$  %) in Fig. 1f, and the maximum difference value between two curves was about 30 %. Anti-correlation between the NPF and RH can be identified at the summit of Mt. Tai, similar to the results in Beijing, Nanjing, Hong Kong, and Mt. Huang (An et al., 2015; Wang et al., 2014a; Shen et al., 2016; Zhang et al., 2016; Guo et al., 2012). The actual role of RH on the NPF still has controversies so far. Hamed et al. (2011) indicated that the RH affected the source of the NPF via decreasing solar radiation under the high RH condition. In addition, increasing CS (sink) under the high RH condition has also been suggested to explain the negative effect of RH (Hamed et al., 2011; Guo et al., 2012).

A wide range of wind speed was on NPF days at the summit of Mt. Tai, varying from  $0.2 \text{ m s}^{-1}$  to  $7.8 \text{ m s}^{-1}$ . Compared with non-NPF days, the wind direction on NPF days had a narrow range in west-southwest ( $250^\circ$ – $300^\circ$ ) and east-southeast ( $85^\circ$ – $110^\circ$ ) (Fig. 5a). These directions corresponded to an elevated  $\text{SO}_2$  concentration, especially in the wind directions between  $270^\circ$  to  $300^\circ$ . This suggests that the gas point sources from the above two directions can increase the probability of the NPF occurrence in a certain extent. The accumulation mode particles showed a high level of concentration when the wind came from the northeast ( $30^\circ$ – $90^\circ$ ), which could, at least partly, explain the less occurrence of NPF from the northeast direction. The nucleation and Aitken modes particles were almost evenly distributed in each wind direction if there was not the NPF event, indicating that there were few local particle sources for the nucleation and Aitken modes particles around the observation site.

To characterize the origins of air masses, air mass backward trajectories for 72 h at 6:00 LT at 1535 m ASL were simulated using the HYSPLIT model developed by the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory. Figure 6a illustrates the air mass backward trajectories of all the NPF days, and Fig. 6b is for all the non-NPF days.

The air mass backward trajectories were mainly classified into three categories: continental air masses, local air masses and maritime air masses. The majority of transport pathways during NPF days were continental air masses, which accounted

for 80 % of the total air masses. Continental air masses on NPF days mainly came from the northwest direction, and they largely originated from Siberia and passed over Mongolia, Inner Mongolia, Shanxi Province, Hebei Province, and Beijing with the long movements. Four-fifths of the continental air masses on NPF days passed through the polluted areas (Beijing, Hebei Province, Shanxi Province, Henan Province, and Shaanxi Province) before reaching the observation site. In contrast, only two-fifths of continental air masses on non-NPF days passed through above polluted areas, and the others were either coming from the south (cleaner part of China) or transporting over Bohai Sea (B-S) and Yellow Sea (Y-S). It seems plausible that air masses going through polluted areas could increase the occurrence of NPF, because transported air masses brought high level of anthropogenic precursors from the polluted areas. In addition, the local and maritime backward trajectories on NPF days accounted for 8 % and 12 % of the total trajectories, respectively (local air masses were 12 % and maritime air masses were 25 % on all non-NPF days). The local air masses were mainly from surroundings such as Jinan, Nanjing, Zhengzhou with short pathways, whereas the maritime air masses originated over the Bohai Sea, Yellow Sea and East China Sea (E-S). Comparison between the Fig. 6a and Fig. 6b found that there were less proportion of maritime air masses and shorter routes over ocean areas on NPF days, suggesting that maritime air masses might not be the favorable conditions for the occurrence of NPF at the summit of Mt. Tai.

### 3.3 Typical NPF events during hazy episodes

In Sect. 3.2, it mainly investigated the favorable conditions for the occurrence of NPF at the summit of Mt. Tai. In fact, the NPF events could also be observed under some specific conditions, such as hazy episodes. According to the Chinese Meteorological Administration, the hazy episode can be identified when the atmospheric visibility is less than 10 km and the RH is less than 80 % simultaneously. In the atmosphere, hazy episodes are always accompanied with elevated  $PM_{2.5}$  concentrations. The higher  $PM_{2.5}$  concentration may mean increased particle loadings, which suppresses the nucleation to a great extent. Indeed, the hourly average  $PM_{2.5}$  concentrations on non-NPF days ( $\sim 26 \mu\text{g m}^{-3}$ , 6:00-9:00 LT) were always higher than the corresponding values on NPF days ( $\sim 20 \mu\text{g m}^{-3}$ , 6:00-9:00 LT) before 9:00 LT. Therefore, an intensive investigation of NPF events occurring during hazy episodes (named as NPF hazy events below) is very important.

In this study, four NPF hazy events (26 July 2014, 17 October 2014, 18 October 2014, and 11 November 2014) were observed during the three campaigns, and the related parameters on four days (named as NPF hazy days below) were shown in Table 4. The  $PM_{2.5}$  concentrations on NPF hazy days were significantly higher than the average of all NPF days observed at the summit of Mt. Tai, but potential high precursor concentrations might exist simultaneously because of the high level of  $SO_2$ ,  $O_3$  and CO concentrations. The temperature and RH were in accordance with the corresponding climates, but wind speed of the each NPF hazy day varied a lot. It was noteworthy that the start time of NPF hazy events was around noon, an obvious time delay compared with the general start time of 8:00-11:00 LT (seen in Sect. 3.1). This phenomenon might suggest that the enhanced solar radiation at noon could be an important factor for the NPF hazy events. In order to further

explore the characteristics of NPF hazy events, one NPF event on 11 November 2014 was selected for case study. The time series of the particle size distribution, meteorological parameters, trace gases, chemical composition and mass concentration of PM<sub>2.5</sub> are illustrated in Fig. 7.

On 11 November 2014, the NPF event was clearly observed at about 11:30 LT. The visibility of this day was always in a low level, and fog episode dispersed with the RH decreasing. It had been hazy episode before the NPF event occurred, and thus the day was considered as NPF hazy day. The highest temperature was about 7 °C and lowest RH was 66 % at noon. At about 10:00 LT, the obvious increase in SO<sub>2</sub> concentration (Fig. 7d), decrease in PM<sub>2.5</sub> concentration and change in chemical composition of PM<sub>2.5</sub> (Fig. 7e) were observed simultaneously. The observation site is located on the mountain top without any stationary source nearby, and above changes therefore may suggest that there was another polluted air mass transported to observation site, contributing to the NPF to a certain extent. The average mass concentrations of sulfate, ammonium, nitrate, OC and EC were 4.1, 4.6, 4.1, 7.6 and 0.7 μg m<sup>-3</sup> during 6:00-9:00 LT, respectively. In contrast, their average concentrations were 13.1, 11.2, 11.1, 10.0 and 0.2 μg m<sup>-3</sup> in order during 11:00-18:00 LT. The ratio of sulfate increased a lot before and after 10:00 LT, suggesting that the new air mass might be S (SO<sub>2</sub>)-rich.

Air mass backward trajectories for 72 h at 1535 m ASL at 14:00, 12:00, 10:00, 8:00 and 6:00 LT on 11 November 2014 are shown in Fig.8. Fig.8 illustrated that the origin of transported air mass moved from eastern China (Jiangsu and Anhui Provinces, line A and B) to western China (line C, D and E) at about 10:00 LT, in agreement with above analysis in Fig. 7. The latter air mass backward trajectories (line C, D and E) passed through the heavily polluted areas, such as Shanxi and Shaanxi Provinces, before reaching the observation site, leading to the potential increase of precursor concentrations.

#### 4. Conclusions

Field observations of the NPF at the mountain-top sites are scarce in China and its related researches could significantly contribute to atmospheric aerosol pollution control. A comprehensive investigation of the NPF was conducted at the summit of Mt. Tai (1534 m ASL), eastern China, from 25 July to 24 August 2014 (I), 21 September to 9 December 2014 (II) and 16 June to 7 August 2015 (III), using two types of size distribution instruments (NAIS and WPS) and including two trace gases, multiple meteorological parameters, mass concentration and chemical composition of PM<sub>2.5</sub>. During the 164 days, 66 NPF events were identified based on the particle size distributions, giving an occurrence frequency of 40 %. Most of the NPF events were initiated at about 8:00-11:00 LT, in agreement with the intensive photochemical activities. The  $J_3$ ,  $J_{3-20}$  and growth rates were in the range of 0.82-25.04 cm<sup>-3</sup> s<sup>-1</sup>, 1.10-57.43 cm<sup>-3</sup> s<sup>-1</sup> and 0.58-7.76 nm h<sup>-1</sup>, respectively. In comparison with other studies in China,  $J_3$  at the summit of Mt. Tai was slightly larger than some rural/suburban environments, which probably was associated with the intensive air mass transport in eastern China and enhanced solar radiation at the summit of Mt. Tai. Instead, growth rates showed the lower values compared with some rural/suburban/urban environments of China,

suggesting insufficient vapors related to growth rate on Mt. Tai.

The CS on NPF days at the summit of Mt. Tai was  $0.1 \times 10^{-2}$ - $28.4 \times 10^{-2} \text{ s}^{-1}$ , much lower than other urban reports in China. The NPF events were in favor to occur in lower CS,  $\text{O}_3$  concentration, temperature and RH conditions, whereas the elevated  $\text{SO}_2$  concentration was observed on NPF days. Without consideration the errors of solar radiation, the average sulfuric acid proxy concentration (6:00-9:00 LT) on NPF days ranged from  $0.52 \times 10^6 \text{ cm}^{-3}$  to  $25.7 \times 10^6 \text{ cm}^{-3}$ . The role of sulfuric acid concentration on the NPF onset might be uncertain because of no evident superiority in sulfuric acid proxy concentration on NPF days in the early morning. The NPF events were more common when the wind was from the west-southwest and east-southeast directions. The majority of air mass backward trajectories on NPF days came from the northwest, and the air masses going through the polluted areas can increase the occurrence of NPF at the summit of Mt. Tai.

Four NPF events were observed during hazy episodes, and their delayed start time might imply that the enhanced solar radiation at noon made an important role on the occurrence of NPF hazy events. Elevated  $\text{PM}_{2.5}$  and trace gases concentrations might be observed on NPF hazy days. As a case study on 11 November 2014, the NPF might be caused by enhanced solar radiation at noon and changing air mass transport which brought the potential high precursor concentration.

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## Figure and Table captions

**Fig. 1.** Average diurnal variations of the condensation sink, sulfuric acid proxy, trace gases (SO<sub>2</sub> and O<sub>3</sub>), and meteorological conditions (T and RH) during NPF days and non-NPF days over all the campaigns at the summit of Mt. Tai.

**Fig. 2.** The relationships between the sulfuric acid proxy, condensation sink and particle number concentration of 3-6 nm ( $N_{3-6}$ ) in the early morning (6:00-9:00 LT) on NPF days (a) and non-NPF days (b). The data are hourly averages, and the spot color represents the values of  $N_{3-6}$  (cm<sup>-3</sup>).

**Fig. 3.** The particle number concentration of 3-6 nm ( $N_{3-6}$ , blue) and sulfuric acid concentration ([H<sub>2</sub>SO<sub>4</sub>], red) on 14 October 2014, fitting a good correlation ( $R^2 = 0.975$ ) during NPF event (6:00-14:00 LT).

**Fig. 4.** Time series during 10 November-9 December 2014 and the shaded areas are NPF days: (a) contour plot of the particle number size distributions using NAIS data; (b) sulfur dioxide (blue) and ozone (red); (c) meteorological parameters, including wind speed (cyan), wind direction (green), temperature (magenta) and relative humidity (earth yellow).

**Fig. 5.** Wind rose plots of all the NPF days (a) and non-NPF days (b), and the wind speed and wind direction between 07:00 and 11:00 LT are included. Length of each spoke on the circle represents the probability of wind coming from a particular direction at the certain range of wind speed.

**Fig. 6.** Air mass back trajectories for 72 h at 6:00 LT at 1535 m ASL on all the NPF days (a) and non-NPF days (b)

**Fig. 7.** Time series of the particle size distributions, meteorological parameters, trace gases, chemical composition and mass concentration of PM<sub>2.5</sub> on 11 November 2014.

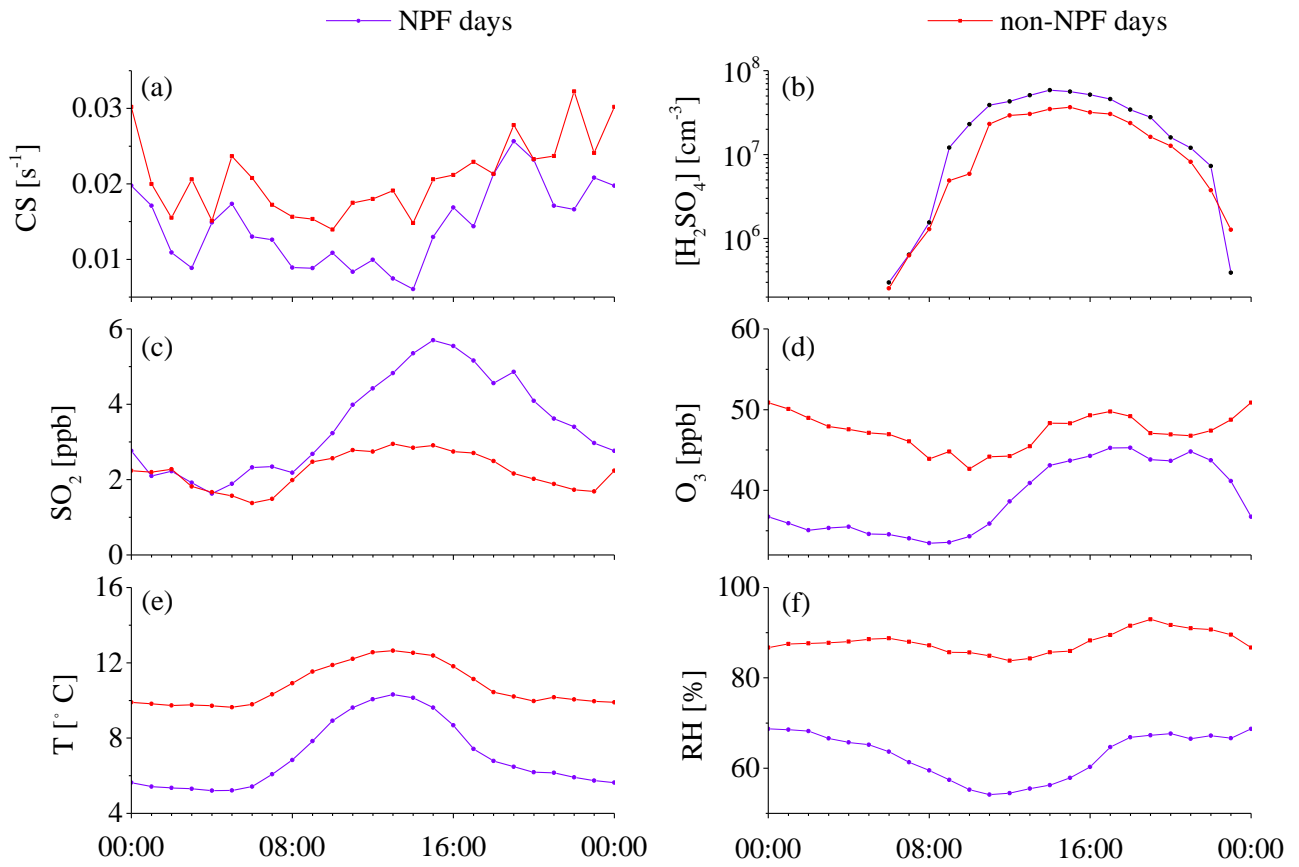
**Fig. 8.** Air mass backward trajectories for 72 h at 1535 m ASL at 6:00 (A), 8:00 (B), 10:00 (C), 12:00 (D) and 14:00 LT (E) on 11 November 2014.

**Table 1.** The calculated parameters of all the NPF events during three campaigns, the minimums and maximums are marked in blue and red, respectively.

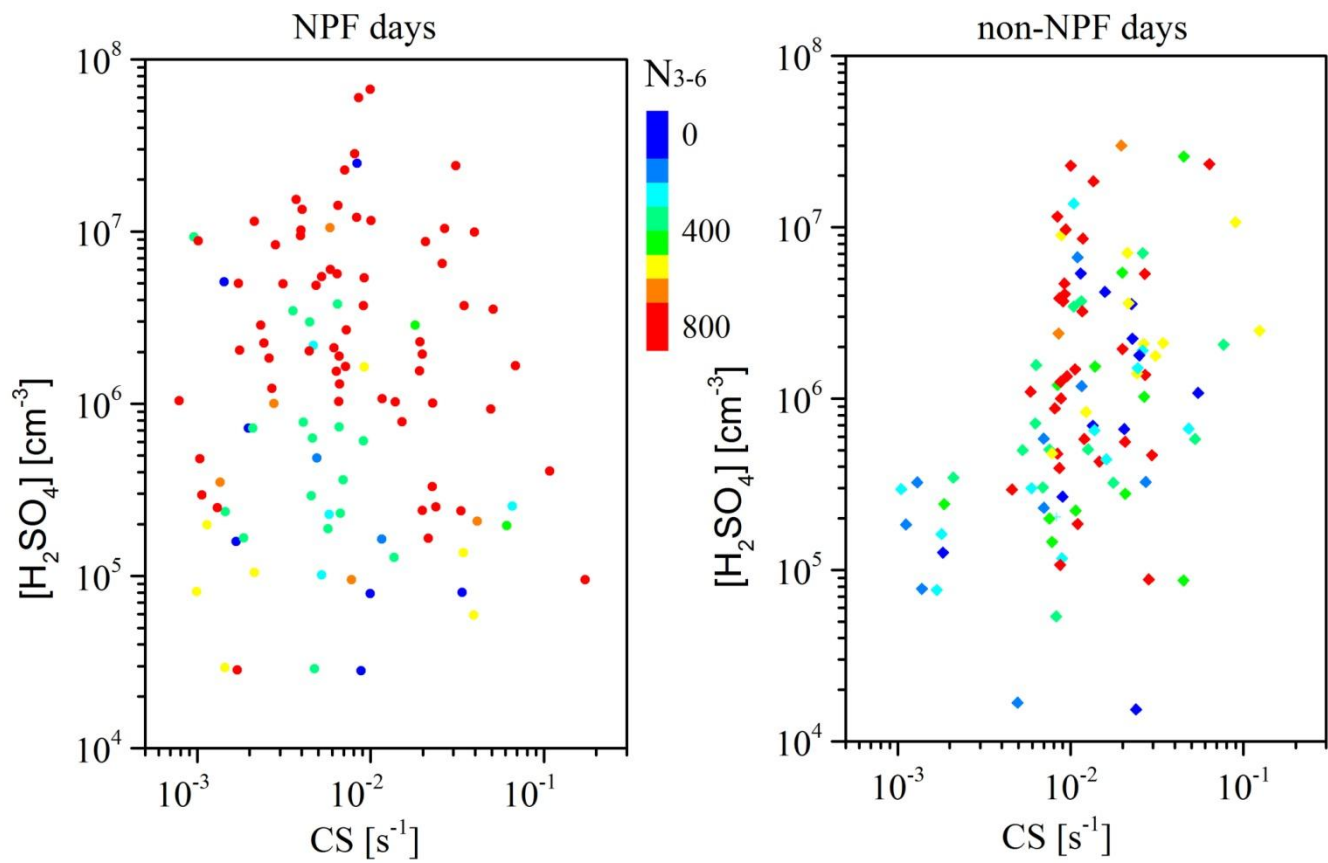
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**Table 3.** The NPF characteristics comparison between Mt. Tai and other typical researches in China

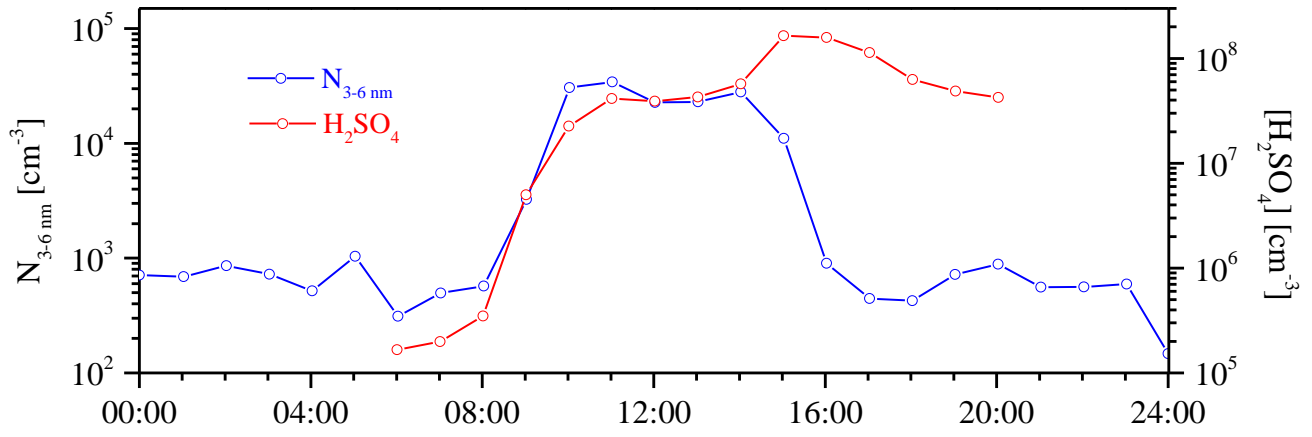
**Table 4.** The parameters on four NPF hazy days and the averages of all the NPF days during daytime (6:00-18:00 LT)



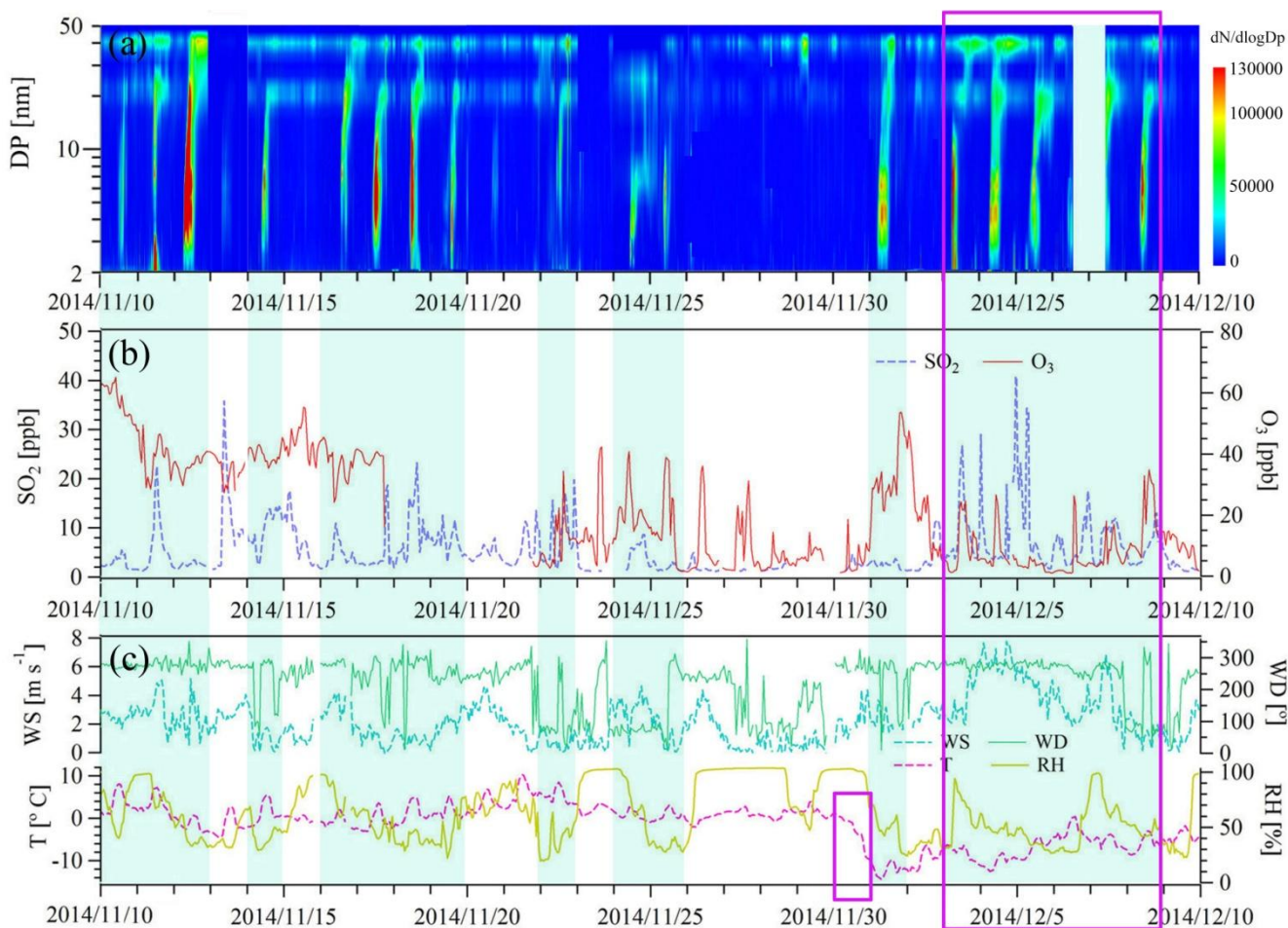
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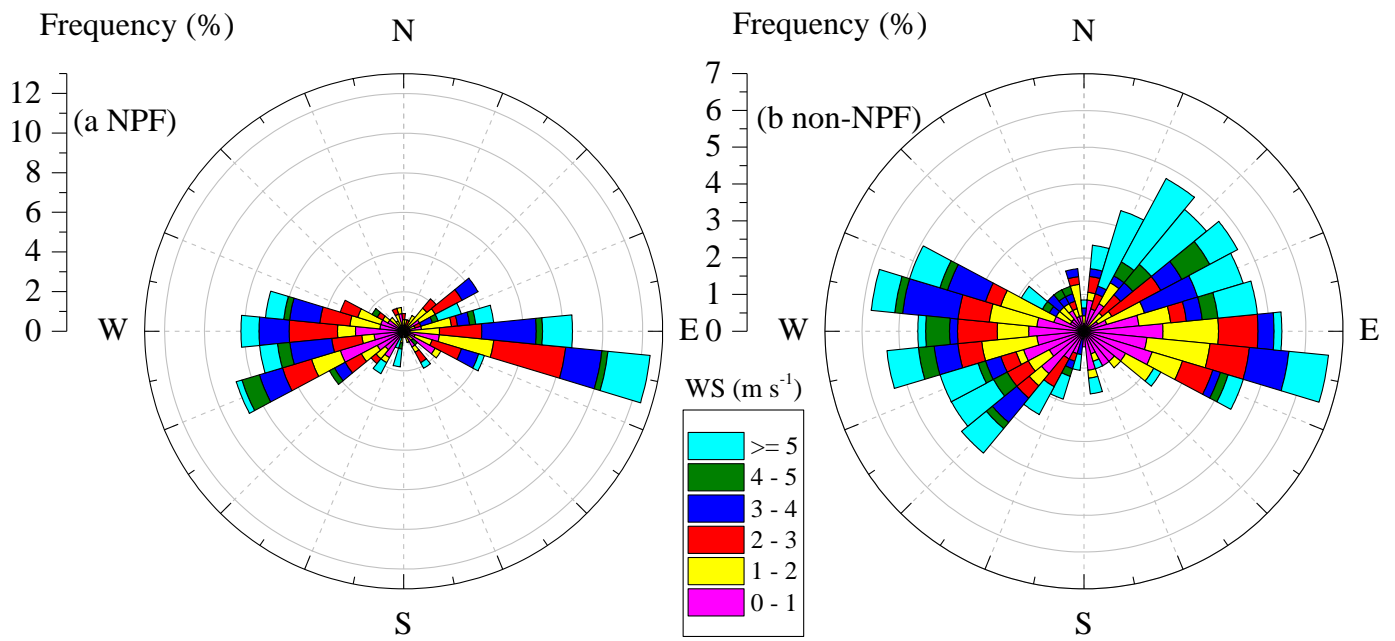


**Fig. 3.** The particle number concentration of 3-6 nm ( $N_{3-6}$ , blue) and sulfuric acid proxy concentration ( $[\text{H}_2\text{SO}_4]$ , red) on 14 October 2014, fitting a good correlation ( $R^2 = 0.975$ ) during NPF event (6:00-14:00 LT).



**Fig. 4.** Time series during 10 November-9 December 2014 and the shaded areas are NPF days: (a) contour plot of the particle number size distributions using NAIS data; (b) sulfur dioxide (blue) and ozone (red); (c) meteorological parameters, including wind speed (cyan), wind direction (green), temperature (magenta) and relative humidity (earth yellow)

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**Fig. 5.** Wind rose plots of all the NPF days (a) and non-NPF days (b), and the wind speed and wind direction between 07:00 and 11:00 LT are included. Length of each spoke on the circle represents the probability of wind coming from a particular direction at the certain range of wind speed.



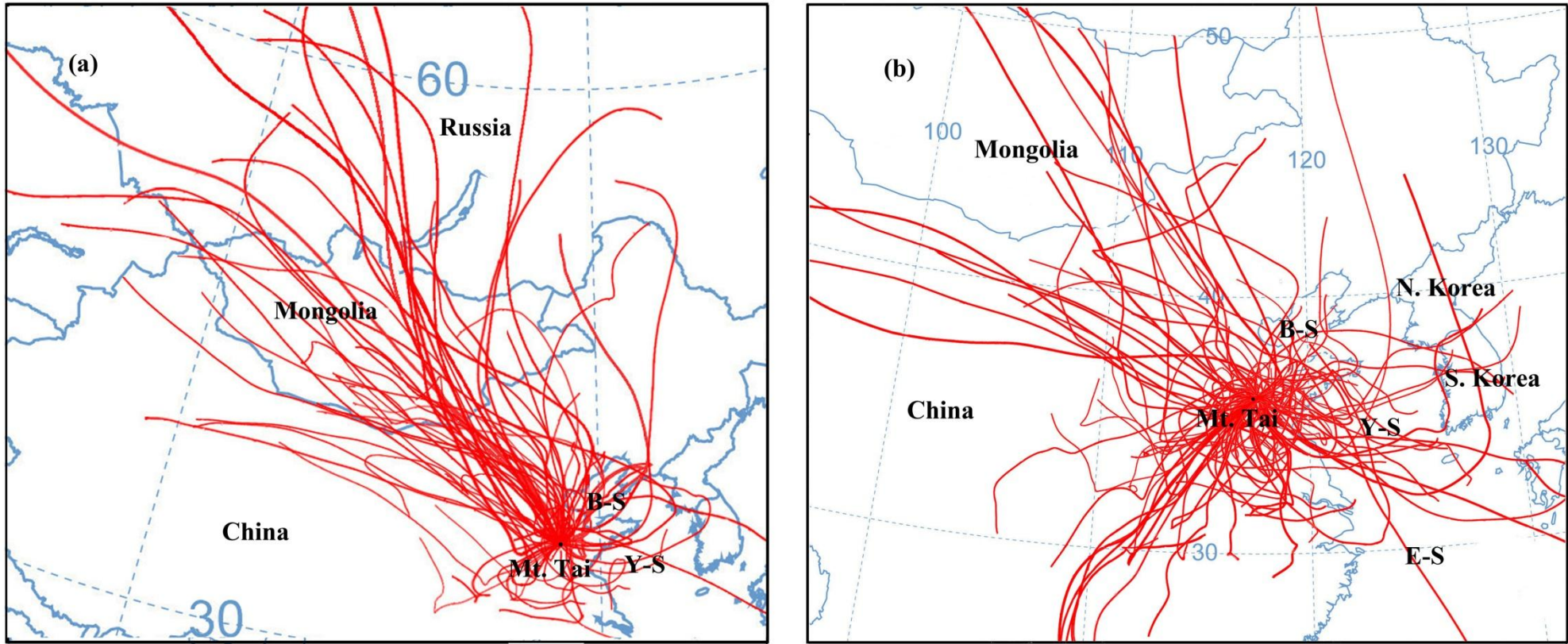
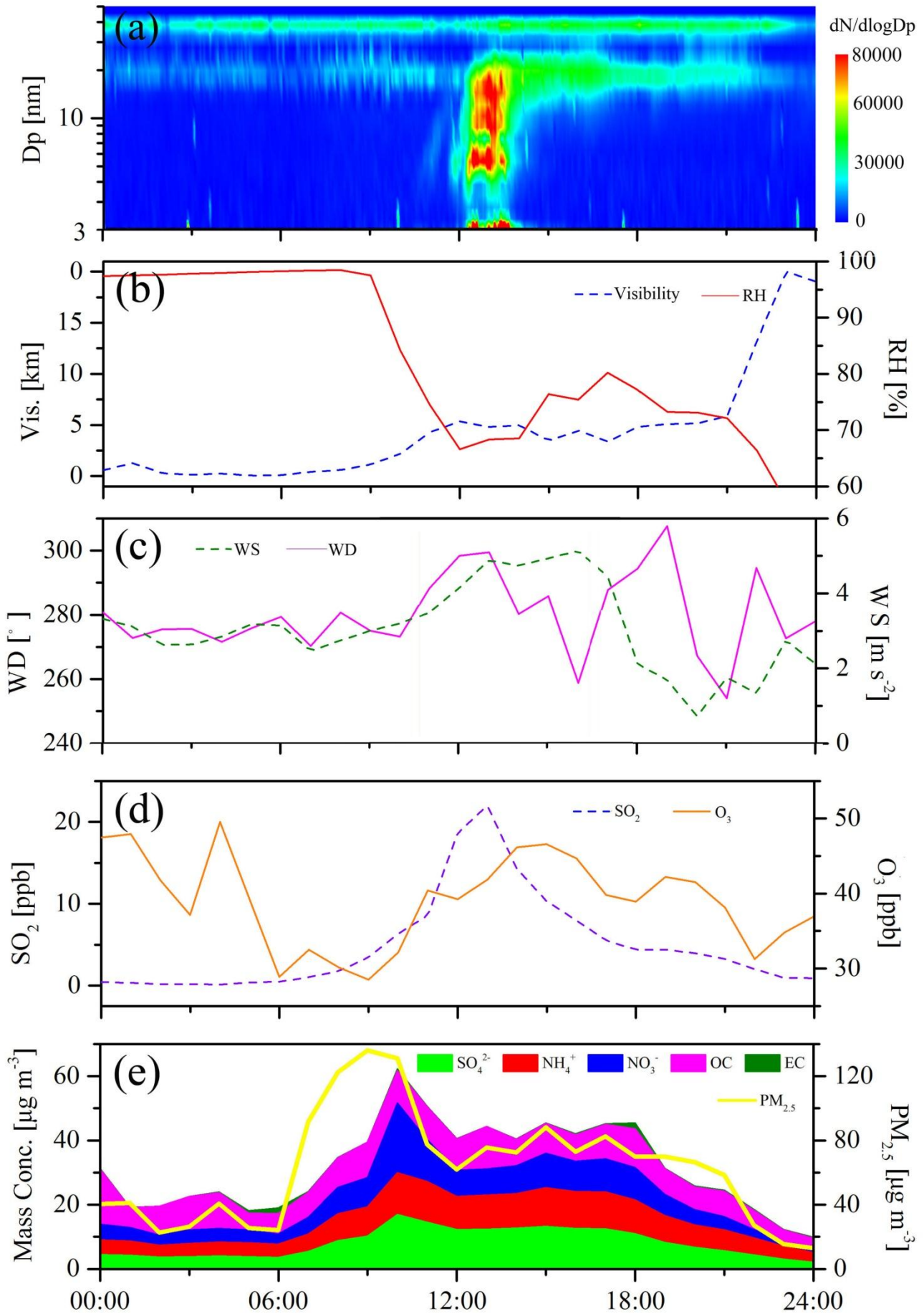
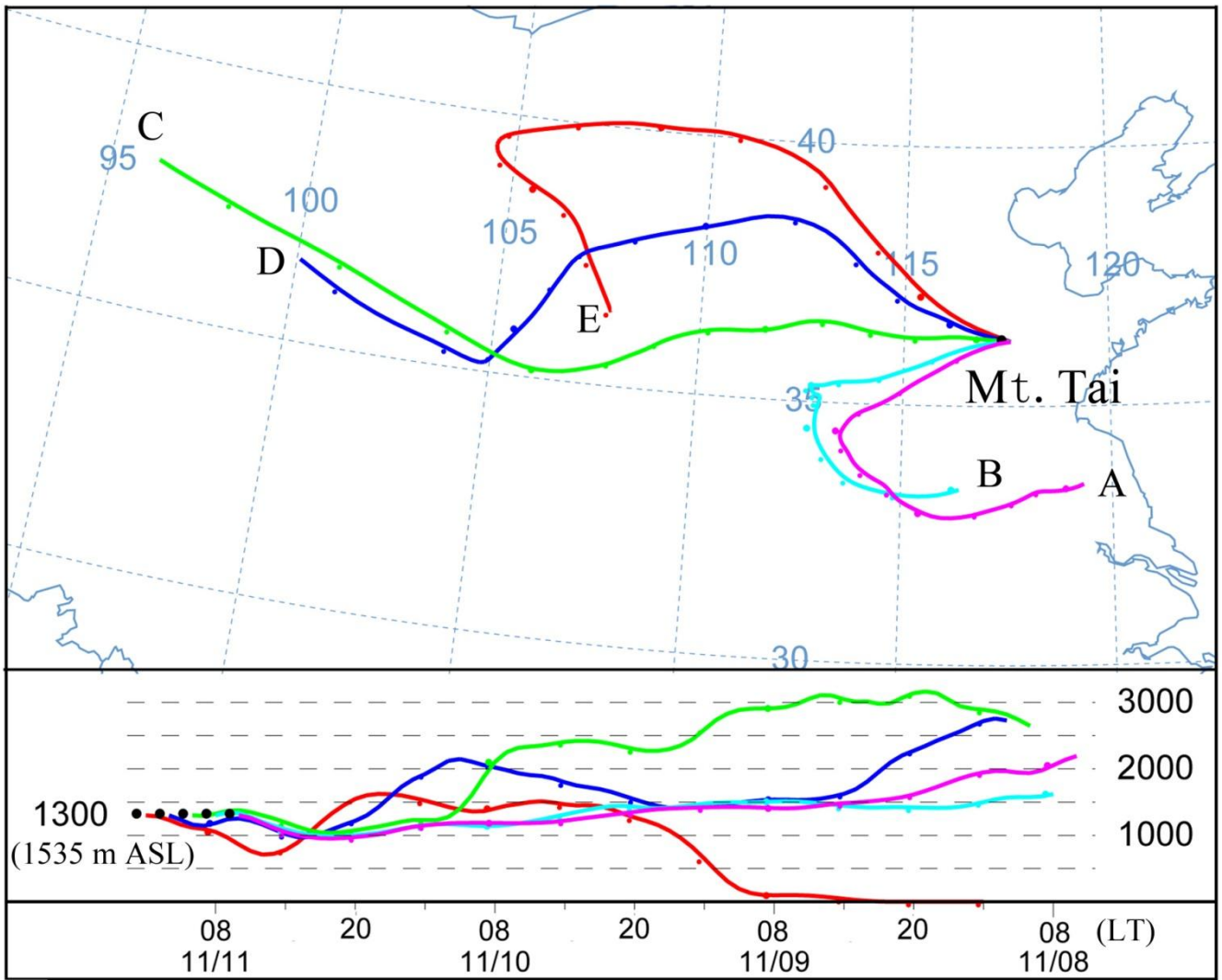


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**Fig. 7.** Time series of the particle size distribution, meteorological parameters, trace gases, chemical composition and mass concentration of  $PM_{2.5}$  on 11 November 2014.



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**Table 1.** The calculated parameters of all the NPF events during three campaigns, the minimums and maximums are marked in blue and red, respectively.

Campaign	Date	$J_3$	$J_{3-20}$	GR	CS	[H <sub>2</sub> SO <sub>4</sub> ]	SO <sub>2</sub>	O <sub>3</sub>	Date	$J_3$	$J_{3-20}$	GR	CS	[H <sub>2</sub> SO <sub>4</sub> ]	SO <sub>2</sub>	O <sub>3</sub>
		cm <sup>-3</sup> s <sup>-1</sup>	cm <sup>-3</sup> s <sup>-1</sup>	nm h <sup>-1</sup>	10 <sup>-2</sup> s <sup>-1</sup>	10 <sup>6</sup> cm <sup>-3</sup>	ppb	ppb		cm <sup>-3</sup> s <sup>-1</sup>	cm <sup>-3</sup> s <sup>-1</sup>	nm h <sup>-1</sup>	10 <sup>-2</sup> s <sup>-1</sup>	10 <sup>6</sup> cm <sup>-3</sup>	ppb	ppb
I	26-Jul-14	3.34	13.32	1.54	0.5-3.1	N/A	N/A	56±7	8-Aug-14	2.61	16.30	1.15	1.8-15.9	2.87	2.7±2.3	65±10
	27-Jul-14	0.94	6.12	1.55	0.6-2.1	N/A	2.4±2.0	56±9	11-Aug-1	N/A	1.44	1.71	0.2-6.9	3.81	0.6±0.5	56±6
	2-Aug-14	N/A	3.60	3.81	0.1-8.7	N/A	N/A	22±13	12-Aug-1	N/A	40.13	3.69	0.8-7.4	22.1	10.8±7.9	79±16
	3-Aug-14	N/A	1.69	<b>7.76</b>	0.1-26.9	N/A	1.5±1.2	35±17	15-Aug-1	2.75	4.52	3.00	N/A	N/A	1.0±1.0	63±12
	6-Aug-14	23.90	52.54	5.78	0.2-1.9	N/A	4.8±2.6	50±5	20-Aug-1	<b>0.82</b>	1.33	4.80	0.6-2.7	9.25	5.2±4.3	72±8
	7-Aug-14	N/A	10.90	1.45	0.1-24.8	N/A	1.5±1.7	56±4								
II	22-Sep-14	0.99	1.58	1.12	0.7-9.0	7.74	5.7±1.7	76±8	4-Nov-14	7.73	15.06	0.77	0.6-1.5	5.39	6.3±1.2	56±3
	29-Sep-14	16.63	54.97	1.13	0.7-21.1	2.43	1.4±1.1	48±5	6-Nov-14	0.98	<b>1.10</b>	1.45	0.1-1.2	2.02	0.6±0.5	13±5
	30-Sep-14	7.94	20.61	1.62	0.5-28.4	1.09	1.9±0.5	2±1	7-Nov-14	1.40	5.59	1.26	0.1-2.0	0.93	0.4±0.3	23±11
	2-Oct-14	5.16	12.02	1.15	0.2-6.6	1.98	1.6±1.7	41±4	8-Nov-14	9.40	13.53	2.95	0.4-2.3	6.71	4.1±2.6	38±3
	3-Oct-14	5.44	14.07	1.50	0.1-4.6	1.34	0.7±0.7	42±3	10-Nov-1	1.02	2.17	3.41	N/A	N/A	2.3±0.7	60±4
	5-Oct-14	8.13	26.41	2.00	0.5-6.9	<b>0.52</b>	0.2±0.1	11±5	11-Nov-1	3.29	14.42	2.52	N/A	N/A	7.8±8.3	34±5
	6-Oct-14	2.54	8.94	0.78	0.4-3.4	0.53	0.3±0.4	7±2	12-Nov-1	12.39	26.36	1.09	N/A	N/A	1.6±0.4	35±2
	8-Oct-14	N/A	1.40	0.92	N/A	N/A	5.0±0.6	71±6	14-Nov-1	9.92	13.41	1.55	N/A	N/A	6.1±4.0	40±2
	10-Oct-14	6.54	27.04	1.80	0.2-1.6	4.33	1.8±2.1	54±3	16-Nov-1	5.36	11.06	2.47	0.5-1.2	11.6	5.7±2.9	34±6
	11-Oct-14	9.43	20.37	0.70	0.2-2.2	1.50	3.1±2.5	68±6	17-Nov-1	9.15	16.82	1.10	0.2-0.9	8.38	1.5±0.2	38±2
	13-Oct-14	6.17	26.84	1.64	0.4-2.0	7.43	5.1±1.6	27±8	18-Nov-1	20.45	57.11	0.80	0.1-1.6	4.42	N/A	N/A
	14-Oct-14	10.15	18.11	1.55	0.1-1.4	1.43	0.9±0.8	44±4	19-Nov-1	8.89	15.41	2.51	0.3-2.8	N/A	N/A	N/A
	15-Oct-14	6.76	30.54	2.76	N/A	N/A	10.1±2.9	60±6	22-Nov-1	N/A	11.60	1.23	0.3-5.6	4.18	5.3±4.9	10±8
	16-Oct-14	6.14	36.96	<b>0.58</b>	N/A	N/A	1.5±1.9	41±1	24-Nov-1	11.03	24.51	1.50	N/A	N/A	2.9±2.1	26±10
	17-Oct-14	1.55	5.75	2.12	N/A	N/A	N/A	N/A	25-Nov-1	7.11	10.97	2.02	0.1-2.9	N/A	0.5±0.4	25±13
	18-Oct-14	6.16	22.89	2.03	N/A	N/A	N/A	N/A	1-Dec-14	4.96	7.55	1.77	0.1-0.9	1.22	1.4±0.3	27±4
	21-Oct-14	N/A	1.98	2.36	0.4-1.3	N/A	0.3±0.3	13±5	3-Dec-14	<b>25.04</b>	<b>57.43</b>	1.51	0.4-1.4	1.64	12.9±9.6	13±11
	24-Oct-14	3.64	8.99	1.25	0.5-1.3	1.12	N/A	N/A	4-Dec-14	4.80	10.62	2.69	0.4-1.4	5.48	2.9±0.3	13±9
	25-Oct-14	7.50	14.74	1.60	0.4-1.4	0.89	0.3±0.3	15±11	5-Dec-14	3.34	5.83	1.32	0.2-1.2	0.95	N/A	4±1
	27-Oct-14	7.24	15.49	0.99	N/A	N/A	0.2±0.3	12±4	6-Dec-14	N/A	1.52	1.99	N/A	N/A	2.2±1.2	4±9
28-Oct-14	2.98	13.17	1.21	N/A	N/A	0.2±0.1	5±2	7-Dec-14	N/A	11.64	1.42	N/A	N/A	7.9±2.5	8±5	
2-Nov-14	11.24	17.78	0.72	0.2-0.9	4.97	0.7±0.1	33±3	8-Dec-14	9.78	16.33	1.24	N/A	N/A	2.4±1.4	16±10	
3-Nov-14	8.77	18.57	1.10	0.3-1.3	6.65	4.6±0.6	37±3									
III	16-Jun-15	1.45	9.22	3.98	1.7-4.9	13.6	6.3±0.8	105±7	4-Jul-15	12.55	23.25	0.91	N/A	N/A	5.7±2.9	95±20
	20-Jun-15	6.16	32.50	3.28	0.5-1.8	10.5	2.8±1.6	81±11	8-Jul-15	N/A	8.25	0.93	0.6-2.3	N/A	0.5±0.5	90±16
	21-Jun-15	3.90	6.08	3.44	1.4-3.0	<b>25.7</b>	10.0±2.1	105±15	13-Jul-15	N/A	5.64	1.95	1.0-2.4	1.10	0.1±0.1	110±15
	2-Jul-15	9.61	43.41	1.08	0.2-1.9	N/A	2.2±1.3	69±10	15-Jul-15	N/A	19.72	2.86	0.7-1.6	N/A	0.2±0.1	88±12
	3-Jul-15	5.54	10.86	2.65	0.2-2.1	N/A	5.5±2.5	101±13	25-Jul-15	N/A	19.56	1.88	0.4-1.9	2.41	0.3±0.2	99±9

**Table 2.** Summary of the mean, median, 25th percentile, 75th percentile, minimum, and maximum for the calculated parameters on the basis of the Table 1

	Mean	Minimum	Maximum	25th percentile	Median	75th percentile
$J_3$ ( $\text{cm}^{-3} \text{s}^{-1}$ )	7.10	0.82	25.04	3.31	6.15	9.41
$J_{3-20}$ ( $\text{cm}^{-3} \text{s}^{-1}$ )	16.61	1.10	57.43	6.12	13.47	20.61
GR ( $\text{nm h}^{-1}$ )	1.98	0.58	7.76	1.15	1.55	2.51
CS ( $10^{-2} \text{s}^{-1}$ )	1.4	0.1	28.4	0.5	0.9	1.7
$[\text{H}_2\text{SO}_4]$ ( $10^6 \text{cm}^{-3}$ )	5.23	0.52	25.7	1.28	3.34	7.07
$\text{SO}_2$ (ppb)	3.2	0.1	12.9	0.7	2.2	2.7
$\text{O}_3$ (ppb)	45	2	110	19	41	66

**Table 3.** The NPF characteristics comparison between Mt. Tai and other typical researches in China

Observation site	FR (cm <sup>-3</sup> s <sup>-1</sup> )	GR (nm h <sup>-1</sup> )	Freq.	Data	Air mass style	Ref.
Mt. Tai	7.10±5.39 ( $J_3$ )	1.98±1.27 (GR <sub>3-20</sub> )	40 %	Jul-Dec 2014 & Jun-Aug 2015	Mountain (1534 m ASL)	This study
Mt. Tai Mo Shan	0.97-10.2 ( $J_{5,5}$ )	1.5-8.4 (GR <sub>5,5-25</sub> )	33 %	Oct-Nov 2010	Mountain (640 m ASL )	Guo et al. (2012)
Mt. Huang	0.09-0.30 ( $J_{10}$ )	1.42-4.53 (GR <sub>10-20</sub> )	37 %	Apr-Jul 2008	Mountain (1840 m ASL )	Zhang et al. (2016)
Mt. Huang		2.29-4.27 (GR <sub>10-15</sub> )	18 %	Sep-Oct 2012	Mountain (869 m ASL )	Hao et al. (2015)
Mt. Daban		0.8-3.2	79 %	Sep-Oct 2013	Mountain (3295 m ASL )	Du et al. (2015)
South Yellow Sea & East China Sea	0.3-15.2 ( $J_{5,6-30}$ )	2.5-5.0	16 %	Oct-Nov 2011 & Nov 2012	Marine	Liu et al. (2014)
Backgarden	2.4-4.0 ( $J_{3-25}$ )	4.0-22.7 (GR <sub>3-25</sub> )	25 %	Jul 2006	Rural	Yue et al. (2013)
Nanjing	2.6 ( $J_6$ )	10.4 (GR <sub>6-30</sub> )	44 %	Dec 2011-Nov 2013	Suburban	Qi et al. (2015)
Lanzhou		1.2-16.9 (GR <sub>10-20</sub> )	33 %	Jun-Jul 2006	Suburban	Gao et al. (2012)
Xinken	0.5-5.2 ( $J_{3-20}$ )	2.2-19.8 (GR <sub>3-20</sub> )	26 %	Oct-Nov 2004	Suburban	Liu et al. (2008)
Shanghai	2.3-19.2 ( $J_3$ )	1.9-38.3 (GR <sub>7-20</sub> )	21 %	Nov 2013-Jan 2014	Urban	Xiao et al. (2015)
Nanjing	1.6-6.7 ( $J_{10-25}$ )	5.6-9.6 (GR <sub>10-25</sub> )	40 %	Jul-Aug 2012	Urban	An et al. (2015)
Beijing	5.0-44.9 ( $J_3$ )	1.86-6.7 (GR <sub>7-30</sub> )	26 %	Jul-Sep 2008	Urban	Wang et al. (2015)
Lanzhou	0.2-6.2 ( $J_{14,6-25}$ )	2.6-12.3 (GR <sub>14,6-25</sub> )	34 %	Aug-Nov 2014	Urban	Zhang et al. (2017)
Qingdao	13.3 ( $J_{5,6-30}$ )	2.0-10.2	41 %	Apr-May 2010	Urban	Zhu et al. (2014)
Hong Kong	1.9 ( $J_{5,5}$ )	3.7-8.3 (GR <sub>5,5-10</sub> )	23 %	Dec 2010-Jan 2011	Urban	Wang et al. (2014a)

**Table 4.** The parameters on four NPF hazy days and the averages of all the NPF days during daytime (6:00-18:00 LT)

	T (°C)	RH (%)	WS (m s <sup>-1</sup> )	SO <sub>2</sub> (ppb)	O <sub>3</sub> (ppb)	NO <sub>2</sub> (ppb)	CO (ppb)	PM <sub>2.5</sub> (µg m <sup>-3</sup> )
Averages	5.4	54	2.1	3.9	31	10.5	759	31
26-Jul-14	19.6±2.1	67±10	0.7±0.4	5.4±3.4	65±12	N/A	N/A	42±3
17-Oct-14	12.8±2.6	59±11	2.7±0.4	N/A	N/A	10.0±4.2	827±256	121±97
18-Oct-14	14.0±2.0	55±8	1.5±0.4	N/A	N/A	7.1±1.9	757±40	49±18
11-Nov-14	4.8±1.9	82±12	3.7±1.1	8.1±6.7	38±6	11.9±3.1	1641±284	84±30