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Title: Vertical profiles of trace gas and aerosol properties over the Eastern North Atlantic: Variations with season and synoptic condition

1. In this paper an in-depth assessment of the vertical heterogeneity of aerosols over this region is presented. The authors must be commended to present such a thorough investigation of several factors influencing the formation and growth atmospheric particulates at different altitudes. Detailed comments related to scientific, technical and language issues are indicated in the annotated PDF file. In general, the paper is well-written, especially from Section 3.3 onwards. There are, however, instances of incoherent writing and discussions as highlighted in the annotated PDF file. There are also some speculative explanations of observations, which are, however, plausible.

Response: We thank the reviewer for the positive feedback. We have thoroughly revised the manuscript based on the comments provided. The point-to-point responses can be found as follows.

2. Line 25: Do you mean in general or only at ground level?

Response: We mean in general. The sentence has been corrected.

3. Line 29: Not exactly sure what you mean here? Condensation being a sink for smaller particles growing into larger particles?

Response: The reduced condensation sink can lead to stronger NPF. The sentence has been corrected into

“..., a result of the stronger new particle formation (NPF) events due to the reduced condensation sink in spring.”

4. Line 32: Is this considered considerable?

Response: We have removed “considerable” and reworded the sentence as

“Through the vertical profiles of aerosol properties, we observed NPF events in the upper BL during 7 out of 38 research flights, where the newly formed particles continue to grow as they are mixed down to the surface.”

5. Line 46 and Line 50: Should be defined in main text.

Response: The abbreviations have been defined.

6. Line 50: Not sure whether this definition of NPF is required.

Response: Since this is an overview of the aerosol measurements during the research campaign, we feel it is necessary to include the definition of NPF to accommodate readers with different backgrounds.

7. Line 54: sulphate

Response: Changed.

8. Line 65: Please define in main text.

Response: The definition has been added.

9. Line 66: Do not like this word.

Response: The word has been replaced by “study.”

10. Line 68: What schemes? In the model or actual formation mechanisms?

Response: In the model. This has been clarified in the manuscript.

11. Line 68: Maybe give a reference here.

Response: Reference is added.

12. Line 69: Not exactly sure what is meant here.

Response: The sentence has been rephrased as

“Apart from newly formed particles, vertically compact aerosol layers, which were traced back to forest fires in East Asia, were observed at altitudes over 3000 m at the SGP.”

13. Line 70: ??

Response: The definition is now added to the main text.

14. Line 85: I do agree with these type of made up abbreviated terms.

Response: Understood.

15. Line 86: I will just say “the spring campaign”

Response: Corrected.

16. Line 89: Nothing mentioned about horizontal coverage.

Response: The horizontal coverage is now added. The sentence is now reworded as

“The Gulfstream-1 (G-1) aircraft (Schmid et al., 2014) was deployed to examine the spatial and temporal variation of aerosol properties over the SGP observatory (36°36’26” N, 97°29’16” W), with a horizontal coverage of around 100 km (Fast et al. 2022) and an altitude range of 200 to 3000 m above ground level (AGL).”

17. Line 92: Who mean diurnal. Temporal refer to seasonal and diurnal.

Response: The word “temporal” has been replaced by the word “diurnal.”

18. Line 92: ???

Response: We meant that there were four to six vertical profiles of G-1 measurements in addition to the one departing from the airport and the one arriving at the airport. The departing and arriving sections are excluded from the analysis to avoid interference from the airport and urban pollution. The sentence has been rephrased as

“Each flight consisted of four to six vertical profiles (in addition to those leaving and arriving at the airport), providing the aerosol and corresponding meteorological properties as a function of altitude.”

19. Line 94: ???

Response: The sentence has been rephrased as

“The flight tracks used during the HI-SCALE campaign and the flight track of an example research flight are shown in **Fig. 2.**”

20. *Line 111: Not sure what is meant is this term?*

Response: The term “non-refractory” is assigned to species that evaporate rapidly at 600 °C under vacuum conditions. It is used for discussing aerosol composition measured by the aerosol mass spectrometers. This definition is now added to the manuscript.

21. *Line 112: ??*

Response: Please see the response to the previous comment.

22. *Line 119: In general, this section can be written more coherently.*

Response: Thanks. We have reorganized the structure of this section. Please see our revised manuscript.

23. *Line 120: Are these averages?*

Response: Yes. This information is added to the manuscript.

24. *Line 124: Maybe consider to put in Experimental/Methods section.*

Response: The description of the back trajectory calculation has been shortened.

25. *Line 128: A map indicating vegetation and agricultural coverage, as well as burned areas (e.g. MODIS satellite imagery) would be useful and support your interpretation.*

Response: A satellite imagery showing the terrain and vegetation surrounding the SGP observatory is now included as **Fig. S1**. However, it is challenging to show the burned area in the imagery.

26. *Line 138: Air masses does not originate from a region, it passes over regions. This is very important.*

Response: Thanks. The sentences have been revised.

27. *Line 139: How and why?*

Response: Further discussion has been added as

“Also, the short trajectory from the east reflects slower wind speeds from the forests during the summertime. The longer residence time of air mass and the more abundant fresh volatile organic compounds in the forest region can lead to a higher mass of aerosols and less oxidized organic aerosols during the summer (Liu et al., 2021).”

28. *Line 146: Why?*

Response: The sentence has been rephrased as

“The wildfire-generated biomass burning aerosols in the north and west of SGP may be injected into the FT, and further transported and aged before they arrive at the SGP.”

29. *Line 149: Reference*

Response: The reference has been added.

30. *Line 149: Do you mean in relation to the impact on ground observations?*

Response: Yes. The sentence has been rephrased as

“Long-range transport of aerosols may impact ground observations under such situations (Wang et al., 2006), but the influence of this air mass on ground observations is also largely subject to the coupling between BL and FT.”

31. Line 153: Very long sentence. Can you show the seasonality of fire frequencies? It is important to support your interpretation here.

Response: The seasonality of the fire frequencies can be found in *Fire Season Climatology in by NWCG: Fire Behavior Field Reference Guide, PMS 437, 2021* (<https://www.nwcg.gov/publications/pms437/weather/fire-season-climatology>). The report shows that overall, regions to the north and west of SGP have higher fire frequencies in spring and summer, while the southeast region has a lower fire frequency. The sentence has been rephrased as

“Fire season climatology shows that, in general, there are stronger fire activities in late spring and summer to the north and west of SGP (NWCG, 2021). The stronger fire activities coincide with the warmest and driest conditions, together with wind events and dry lightning potential. There is a much weaker fire activity to the southeast of SGP in late summer, especially from August to September.”

32. Line 158: Not potentially, it does.

Response: The word has been removed.

33. Line 162: What is meant here? Dependency of SO₂ levels or dependency of NPF?

Response: The sentence has been clarified as

“SO₂ can contribute to the occurrence of NPF. At the same time, NPF depends on the concentration of pre-existing aerosols and other meteorological parameters (Section 3.3.1).”

34. Line 171: I think you mentioned that you did not consider measurements in clouds.

Response: Yes, but aqueous chemistry in the cloud droplets can contribute to the size increase of the accumulation mode aerosols.

35. Line 174: Still not sure what is meant here.

Response: The sentence has been rephrased as

“The larger size of the aerosols in the summer also leads to larger condensation and coagulation sinks for nucleating vapors and newly formed particles, suppressing the occurrence of NPF in the summer. As a result, the concentration of aerosols below 30 nm in the BL (e.g., below 1000 m) during spring is higher than that during summer.”

36. Line 175: Maybe include reference.

Response: The reference has been included.

37. Line 177: Term previously defined, i.e. CCN.

Response: The term is previously defined in the abstract only. The abbreviation is introduced in the main text here again.

38. Line 180: “due to” or “through”?

Response: The word is replaced.

39. Line 193: Evidence to support this?

Response: This can be supported by the fact that the mean values of N_{10-20} are outside the 25th to 75th percentile box in the altitude range of 500 and 1000 m. In the spring campaign, there were three NPF events where N_{10-20} values were above 2000 cm^{-3} in the altitude range of 500 and 1000 m, significantly higher than the median values between 400 and 600 cm^{-3} . More discussion has been added as

“This can also be indicated by the mean values of N_{10-20} being outside the 25th to 75th percentile box in the altitude range between 500 and 1000 m (**Fig. 5b**). In the spring campaign, there were three NPF events where N_{10-20} values were above 2000 cm^{-3} in the altitude range of 500 and 1000 m, significantly higher than the median values between 400 and 600 cm^{-3} .”

40. Line 202: A better site description/map will greatly assist in understanding/supporting interpretations.

Response: To assist in supporting the interpretations, we included a satellite image showing the terrain and vegetation surrounding the SGP observatory as **Fig. S1**.

41. Line 216: in the BL?

Response: Yes. The sentence has been rephrased as

“A breakdown of the total aerosol concentration into different size modes shows that, in the BL, N_{10-20} is higher during the spring (**Fig. 5b**), while $N_{>100}$ is higher during the summer (**Fig. 5d**).”

42. Line 216: Evidence?

Response: Please see the response to comment 39.

43. Line 243: Why?

Response: The sentence has been rephrased as

“Since nitrate aerosols are more stable under low temperatures, the higher nitrate concentrations during spring are likely driven by the lower temperature in the spring (Liu et al., 2021; Parworth et al., 2015).”

44. Line 247: Refer to figure.

Response: Figures 3 and S1 are now referred to in the discussion.

45. Line 247: Not sure how this is relevant to your observation? Sulphate is usually important for formation of particulate and organic species for particle growth. Maybe just explain it better.

Response: As Xu et al. (2015) pointed out, isoprene-derived SOA is directly mediated by the abundance of sulfate through chemical reactions among sulfate, organics, and water. These reactions allow the increase in both the sulfate and organics in the summer. The discussion has been added as

“Moreover, the previous study by Xu et al. (2015) showed that the formation of isoprene SOA is mediated by the abundance of sulfate through chemical reactions among sulfate,

organics, and water, which allows the increase of both organics and sulfates in the summer.”

46. Line 251: Usually indicative of fresher plumes.

Response: These organics are more likely caused by fresh biogenic emissions instead of biomass-burning plumes. The study by Liu et al. (2021) analyzing the aerosol mass spectrometer data showed that the biomass-burning organic aerosol (BBOA) factor was not identified during the summer season. This result is also consistent with the low concentrations of BBOA observed in summer at SGP in a previous study (Parworth et al., 2015). More discussion has been added as
“These summertime organics are not generated by biomass-burning plumes as the study by Liu et al. (2021) analyzing the aerosol mass spectrometer data showed that the biomass-burning organic aerosol (BBOA) factor was not identified during the summer season. This result is also consistent with the low concentrations of BBOA observed in summer at SGP in a previous study (Parworth et al., 2015).”

47. Line 253: This means that the aerosol might not even reach SGP.

Response: The sentence has been rephrased. The springtime aerosols are more aged because they have a longer residence time in the atmosphere and different oxidant concentrations, or a combination of both effects. The discussion has been revised as
“According to Liu et al. (2021), the different oxidation levels in the spring and summer are likely due to (1) aerosols in the spring are more aged due to a longer residence time in the atmosphere, potentially different oxidant concentrations, or a combination of both effects.”

48. Line 254: Not sure whether I agree.

Response: More discussion has been added as
“(2) different VOC species could contribute to aerosol formation in spring and summer (e.g., higher monoterpene and isoprene concentrations were observed in the summer), and (3) the more abundant biogenic VOCs in the summer were not transformed into a higher-oxygenated form in the aerosol phase, either due to radical chemistry, oxidants, or their residence time in the atmosphere.”

49. Line 320: I would expect this.

Response: Agreed.

50. Line 338: Due to oxidation to SO₄?

Response: Agreed. More discussion has been added as
“(2) SO₂ was already depleted in the upper BL due to its oxidation to sulfate that promoted the NPF event.

51. Line 375: This seems to be repeating information.

Response: The term “volume-controlled processes” was discussed in the earlier section of the manuscript. However, “coagulation” and “condensation” were not explained. The sentence has been reworded as

“The particles can grow through different dynamic approaches, including coagulation, condensation, and volume-controlled processes.”

52. Line 381: Also, please look at the following paper and cite: Vakkari et al, 2015, Reevaluating the contribution of sulfuric acid and the origin of organic compounds in atmospheric nanoparticle growth, *Geophysical Research Letters*, 42, 10,486–10,493, doi:10.1002/2015GL066459

Response: Thank you. The discussion of this reference is now added to the manuscript as “The aerosol measurement in a South African grazed savannah grassland environment showed that depending on the gaseous precursors and size of the newly formed particles, the aerosol growth can be dominated by either sulfuric acid accompanied by ammonium or organic compounds originating in either biogenic emissions or savannah fires. The contribution of sulfuric acid was larger during the early phases of the growth, but in clean conditions, organic compounds dominated the growth from 1.5 nm up to climatically relevant sizes.”

53. Line 386: Is abbreviated term defined?

Response: Thanks for pointing this out. The term “extremely low volatility organic compounds (ELVOCs)” is now defined in the earlier section of the manuscript.

54. Line 517: average?

Response: Yes, it is the mean concentration. The word has been added.

55. Line 564: You did it only for two seasons. Not full seasonal pattern.

Response: Thanks. The sentence has been revised as “In this study, we present aerosol properties and meteorological parameters characterized onboard the G-1 aircraft during the spring and summer HI-SCALE campaigns. The key processes that drive the aerosol population in the BL are investigated by examining the vertical variation of aerosol properties.”

56. Line 588: Strong word. “Significantly improved understanding” maybe?

Response: The sentence has been revised. Thank you.

We thank the reviewer for his/her comments. This has improved the quality of the manuscript and we look forward to this manuscript being accepted.