Influence of terrestrial and marine air-mass on the constituents and intermixing of bioaerosols over coastal atmosphere

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Response to editor

We thank the editor for the opportunity to revise our manuscript, we appreciate the editor and reviewer very much for their positive and constructive comments and suggestions. We have revised our manuscript according to the reviewer's comments, and the response to the reviewer's comments are in detailed below. We also attach a revised manuscript with tracked changes, and the amendments were marked in red in the revised manuscript.

Response to reviewer

I would like to thank the authors of the manuscript entitled "Influence of terrestrial and marine air-mass on the constituents and intermixing of bioaerosols over coastal atmosphere" for taking the time to respond clearly and in detail to all the comments. I find that the manuscript has been significantly improved and, I believe, deserves to be published in Atmospheric Chemistry and Physics. That being said, I have additional comments that I would appreciate you considering.

Response of the authors:

We greatly appreciate the positive comments and constructive suggestions on our

manuscript. According to your suggestions, we have carefully incorporated them into our paper and made detailed revision based on your comments.

1. L338-347 and answer to the comment 15 of the authors: I am not criticizing the qPCR detection method at all, as it is a widely validated method in the literature. I am simply questioning the semantics used: qPCR does not provide information on the number of cells per m³ of air, but rather on the number of 16S genes, usually expressed as copies per m³ of air. Most bacteria have multiple copies of the 16S gene in their genome, ranging from 1 to 15. In your case, the methodology used (employing E. coli as a standard) allows you to approximate a cellular concentration, but it remains an estimation. I would rather refer to genome copies per m³ of air.

Response of the authors:

We thank the reviewer for the comments and suggestions. QPCR does not quantify the number of cells per m³ of air, but rather the number of 16S genes, usually expressed as copies per m³ of air. Most bacteria have multiple copies of the 16S gene in their genome. We used the data on bacterial rRNA gene copy number from the rrnDB database (https://rrndb.umms.med.umich.edu/) (Stoddard et al., 2015). According to the latest data, the copy number of the 16S rRNA gene in their genome ranges from 1 to 21 with an average value of 5.5.

The number of bacterial cells, calculated based on the mean copy number of 5.5 in the bacterial genome, is an estimate value. The rRNA copies per genome may range from 50 to 100 in filamentous fungi (Rooney and Ward, 2005) and ascomycetes often have relatively smaller genome sizes (Kullman et al., 2005). The copy number of the fungal ITS gene in their genome was 50 in this study.

As suggested by the reviewers, we have changed the unit of bacterial and fungal number to copies per m³ of air. The Figure 2, and Section 3.3 Bacterial and fungal count, have been revised according to the unit of copies per m³ of air.

HICHIGAN MEDICINE		<i>rm</i> DB							The Ribosomal RNA Database Curated by the <u>Schmidt Laboratory</u>
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Figure 1 The estimate of copy number of 16S rRNA genes in domain Bacteria

Reference

Kullman, B., Tamm, H., and Kullman, K.: Fungal genome size database, http://www.zbi.ee/fungal-genomesize, 2005.

Rooney, A. P. and Ward, T. J.: Evolution of a large ribosomal RNA multigene family in filamentous fungi: birth and death of a concerted evolution paradigm, Proc. Natl. Acad. Sci. USA, 102, 5084-5089, https://doi.org/10.1073/pnas.0409689102, 2005.

Stoddard, S. F., Smith, B. J., Hein, R., Roller, B. R. K., and Schmidt, T. M.: rrnDB: improved tools for interpreting rRNA gene abundance in bacteria and archaea and a new foundation for future development, Nucleic Acids Res., 43, D593-D598, https://doi.org/10.1093/nar/gku1201, 2015.

There are also many spelling errors and typos in this manuscript. I suggest a thorough review to correct them all. Here are the ones I noted:

Response of the authors: Thank you for highlighting the spelling errors in our manuscript. We have reviewed and revised the manuscript, and the spelling errors have been corrected.

2. L60: there is a comma missing after "environment".

Response of the authors: We have revised the sentence in Line 59-61.

The geographical and topographical factors, such as terrestrial and marine environments, exhibit significant differences in bioaerosol sources and pollution characteristics.

3. L71: "maybe" should read "may be".

Response of the authors: We have revised the sentence in Line 71-73.

Bioaerosols from the oceans may be influenced by long-distance transport from continental sources, such as plants and human pathogens (Elbert et al., 2007; Sharoni et al., 2015).

4. L74: seems there is a word missing after "marine". Maybe the authors mean "marine

environment or surface"?

Response of the authors: We have revised the sentence in Line 73-76. Studies have shown that the concentration and diversity of bacterial and fungal aerosols from marine environment are typically lower than those derived from continental sources (Cao et al., 2024; Xue et al., 2022; Shi et al., 2022).

5. L104: "costal" should read "coastal".

Response of the authors: We have revised the sentence in Line 104-106.

The coastal aerosols provide the ideal conditions for understanding the mixing processes taking place between natural and anthropogenic air masses from terrestrial and marine.

6. L124: "between January to March" should read "between January and March" or "from January to March".

Response of the authors: We have revised the sentence in Line 124-125.

 $PM_{2.5}$ samples were gathered between January and March, 2018, during the winter heating and spring dust seasons in northern China.

7. L127: there is a comma missing after "utilized".

Response of the authors: We have added the comma in Line 125-129.

Two parallel $PM_{2.5}$ samplers (TH-150C-III, Wuhan Tianhong Instrument Co., Ltd., China) with a particle size of $2.5 \pm 0.2 \mu m$ were utilized, equipped with Quartz membrane for the collection of $PM_{2.5}$, inorganic ions, metal elements, and microorganisms at a flow rate of 100 L min⁻¹.

8. L128: "ion" should read "ions".

Response of the authors: We have revised the sentence in Line 125-129.

Two parallel $PM_{2.5}$ samplers (TH-150C-III, Wuhan Tianhong Instrument Co., Ltd., China) with a particle size of $2.5 \pm 0.2 \mu m$ were utilized, equipped with Quartz membrane for the collection of $PM_{2.5}$, inorganic ions, metal elements, and microorganisms at a flow rate of 100 L min⁻¹.

9. L132: the tilde should be replaced by "to" (7:00 to 18:30).

Response of the authors: We have revised the sentence in Line 129-132.

The Quartz membranes were cauterized in a muffle furnace at 450°C for 6 hours to remove carbonaceous and contaminant materials. Intermittent sampling was used and samples were collected twice a day (7:00 to 18:30 and 19:00 to 06:30 the following day).

10. L161-162: "metallic elementals" should read "metallic elements".

Response of the authors: We have revised the sentence in Line 161-164.

Metallic elements including Al, Fe, Ti, Mn, Co, Ni, Cu, Zn, Ga, Sr, Cd, Sn, Sb, Pb, V, Cr, and As, were extracted using microwave digestion extraction (ETHOS ONE, Milestone), with the concentrations determined by ICP-MS or ICP-OES (Thermo Fisher).

11. L169: "were simulated one-hour intervals" should read "were simulated at one-hour intervals".

Response of the authors: We have revised the sentence in Line 169-170. Backward trajectories were simulated at one-hour intervals and estimated over a 24-hour period.

12. L176: "and mixed air-mass sample" should read "and a mixed air-mass sample". **Response of the authors:** We have revised the sentence in Line 173-178.

In this study, air mass categories were defined that if more than 90% of the masses originated from the ocean it was a typical sample affected by marine airmass; a typical terrestrial air-mass sample if more than 90% originated from the continent; and a mixed air-mass sample if the proportions of terrestrial and marine air masses were similar or accounted for more than 40% of the total air masses in one day.

13. L227: "Bacterial community functional was conducted" should read "Bacterial community functional analysis was conducted".

Response of the authors: We have revised the sentence in Line 235-238.

Bacterial community functional analysis was conducted using FAPROTAX, a manually constructed database that maps prokaryotic taxa to metabolic or other ecological functions, such as sulfur, nitrogen, hydrogen, and carbon cycling (Chen et al., 2022).

14. L228-229: "other ecologically functions" should read "other ecological functions". **Response of the authors:** We have revised the sentence in Line 235-238.

Bacterial community functional analysis was conducted using FAPROTAX, a manually constructed database that maps prokaryotic taxa to metabolic or other ecological functions, such as sulfur, nitrogen, hydrogen, and carbon cycling (Chen et al., 2022).

15. L234: « Pathorotroph » should read « Pathotroph".

Response of the authors: We have revised the sentence in Line 241-242.

Three primary groups are obtained based on the nutritional mode: Pathotroph, Symbiotroph, and Saprotroph.

16. L235: "different airmasses" should read "different air masses".

Response of the authors: We have revised the sentence in Line 243-244. Samples affected by different air masses were examined for intergroup species variability, based on community abundance data.

17. L247-248: "microbial community and various environmental factors" should read "microbial community composition and various environmental factors".

Response of the authors: We have revised the sentence in Line 254-255. *The Mantel analysis was utilized to reveal the correlation between microbial*

community composition and various environmental factors.

18. L257: "During reginal haze pollution" should read "During regional haze pollution". **Response of the authors:** We have revised the sentence in Line 265-267.

During regional haze pollution, the terrestrial air masses primarily influenced Weihai were typically originating from the Beijing-Tianjin-Hebei region and the surrounding areas.

19. L281: "Cold northwestern airmass form the continent" should read "Cold northwestern airmass from the continent".

Response of the authors: We have revised the sentence in Line 289-290.

Cold northwestern airmass from the continent, and marine air masses from east or south, were the primary contributors during pollution mitigation.

20. L288: "northwestern airmass form the continent" should read "northwestern airmass from the continent".

Response of the authors: We have revised the sentence in Line 295-297.

Pollution elimination was initiated with high winds, snowfall influenced by the cold northwestern airmass from the continent and marine air masses from the northeast sea.

21. L302: please replace "succeeded" by "followed".

Response of the authors: We have revised the sentence in Line 307-312.

In marine air-mass samples, a lower concentration of water-soluble ions was observed, with the concentration of $13.01\pm7.43 \ \mu\text{g/m}^3$, $27.94\pm13.61 \ \mu\text{g/m}^3$ and $30.38\pm11.38 \ \mu\text{g/m}^3$ in marine, terrestrial and mixed air masses, respectively. Notably, NO₃⁻ had the highest proportion (26.94%, 6.4%~52.6%), followed by SO₄²⁻ (21.94%, 9.4%~33.4%) and NH₄⁺ (20.26%, 5.8%~35.6%).

22. L309: "with a range that from" should read "that ranged from" or "with a range of".

Response of the authors: We have revised the sentence in Line 318-319.

A high concentration of Na^+ was observed, with a range of $3.15\pm1.69 \ \mu g/m^3$, and accounted for 14.47% of the total water-soluble ions.

23. L310: "ion" should read "ions".

Response of the authors: We have revised the sentence in Line 318-319.

A high concentration of Na⁺ was observed, with a range of $3.15 \pm 1.69 \ \mu g/m^3$, which accounted for 14.47% of the total water-soluble ions.

24. L315: "The concentration of K⁺ 0.24 \pm 0.20 µg/m³" should read "The concentration of K⁺ was 0.24 \pm 0.20 µg/m³".

Response of the authors: We have revised the sentence in Line 323-326.

The concentration of K^+ was $0.24\pm0.20 \ \mu g/m^3$ and $0.26\pm0.10 \ \mu g/m^3$ in the terrestrial and mixed air-mass samples, and was twice as high as those in the marine air-mass samples $(0.11\pm0.05 \ \mu g/m^3)$, which suggested an important contribution from anthropogenic emissions.

25. L324: "coal combination in winter heating" probably did the authors mean "coal combustion"?

Response of the authors: We have revised the sentence in Line 330-332.

Overall, from the composition and concentration of water-soluble ions in $PM_{2.5}$, the coastal city was more affected by sea salt, coal combustion and dust events in early spring.

26. L356: "in both terrestrial and mixed air-mass samples" should read "in both terrestrial and mixed air-masses samples".

Response of the authors: We have revised the sentence in Line 363-364.

Cyanobacteria exhibit a higher concentration in both terrestrial and mixed air-masses samples.

27. L366: please replace "including" by "included".

Response of the authors: We have revised the sentence in Line 374-376. *These bacteria included a series of opportunistic pathogens and were found abundant in terrestrial and mixed air masses samples.*

28. L372: "to the previously studies" should read "to the previous studies". **Response of the authors:** We have revised the sentence in Line 379-381.

The dominant fungal phyla were Ascomycota (77.29%) and Basidiomycota (21.58%), which were similar to the previous studies (Du et al., 2018; Liu et al., 2019; Zeng et al., 2019).

29. L388: I guess that "and automatic compound" should read "and aromatic compound".

Response of the authors: We have revised the sentence in Line 394-395. Marine air-mass samples were enriched with mammal gut bacteria, as well as hydrocarbon and aromatic compound degradation bacteria.

30. L394: "the prevalence of Saprotroph fungi was observed higher in samples" should read "the observed prevalence of Saprotroph fungi was higher in samples".

Response of the authors: We have revised the sentence in Line 400-401. In particular, the observed prevalence of Saprotroph fungi was higher in samples from marine air masses, such as those containing Aspergillus.

31. L406: there is one extra parenthesis.

Response of the authors: We have revised the sentence in Line 412-414. *These include bacteria such as Bacillus, Streptococcus, and Deinococcus (Maki et al., 2010; Park et al., 2018; Qi et al., 2021).*

32. L408: "airmasses" should read "air masses".

Response of the authors: We have revised the sentence in Line 414-415. These microorganisms can adhere to particulate matter during long-distance transport of air masses after being released from their terrestrial habitats.

33. L413: please replace "conducted" by "shown".

Response of the authors: We have revised the sentence in Line 420-421. *Community disparities influenced by terrestrial, marine, and mixed air masses was shown in Table S2, S3, Fig. 3 and Fig. 4.*

34. L415: "air mass" should read "air mass samples".

Response of the authors: We have revised the sentence in Line 421-423. *The Principal Coordinates Analysis (PCoA) revealed distinct clusters corresponding to terrestrial, marine, and mixed air mass samples (Fig. S4).*

35. L467: "automatic compound degradation bacteria" should read "aromatic compound degradation bacteria".

Response of the authors: We have revised the sentence in Line 472-474.

Marine air-mass samples were enriched with mammal gut bacteria, hydrocarbon and aromatic compound degradation bacteria, and undefined Saprotroph fungi.

36. L482: "diverse and abundance microbial populations" should read "diverse and abundant microbial populations".

Response of the authors: We have revised the sentence in Line 487-489.

Air masses transported over long distances from the continent appear to harbor diverse and abundant microbial populations (Kakikawa et al., 2009; Deleon-Rodriguez et al., 2013).

37. L488-490: "...accompanied the dust transportation to the downwind of Asian Dust including the coastal city of Weihai. Influenced by mixed air masses, bacterial community was 490 significantly positively correlated with K^+ (P<0.01) and PM₁₀ (P<0.05)". The sentence is weird.

Response of the authors: We have revised the sentence in Line 491-494.

Dust-borne bacteria, such as Staphylococcus, Delftia, Pseudoalteromonas and Deinococcus, are likely introduced into the atmosphere during Asian Dust events. Most of these bacteria accompany the transportation of dust particles to the downwind coastal city.

38. L491: "showed high positively correlated" should read "showed high positive correlation" or "were positively correlated".

Response of the authors: We have revised the sentence in Line 495-497. Similarly, microbial communities showed high positive correlation with ions from continental sources, such as K^+ , Mg^{2+} , and Ca^{2+} in terrestrial and mixed air mass samples.

39. L511: Replace "Except" » by "Besides"?

Response of the authors: We have revised the sentence in Line 515-517. Besides anthropogenic sources, such as industrial emissions, the origins of Mg^{2+} and Na^+ in coastal regions should also consider the impact of sea salt (Sun et al., 2022).

40. L517: "temperature have" should read "temperature has".

Response of the authors: We have revised the sentence in Line 522-523. *Influenced by mixed air masses, temperature has a greater impact on fungal community, which was positively correlated with Malasseziales and Davidiellaceae.*

41. L519: "marine airmass samples" should read "marine air mass samples".

Response of the authors: We have revised the sentence in Line 523-527.

In marine air mass samples, a positive correlation between air temperature and certain microorganisms (Aerococcus, Cloacibacterium, Sphingobium, Enhydrobacterium, Davidiellaceae, Malasseziales) also indicated that the increase in air temperature in spring favors the survival of airborne microbes (Jones and Harrison, 2004).