1 **Impact assessment of terrestrial and marine air-mass on the**

2 **constituents and intermixing of bioaerosols over coastal atmosphere**

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12 **Abstract**:

13 Coastal environments provide an ideal setting for investigating the intermixing

14 processes between terrestrial and marine aerosols. Fine particulate matter ($PM_{2.5}$)

15 samples collected from a coastal location in Northern China were categorized into

16 terrestrial, marine and mixed air masses. Chemical and biological constituents during

17 the winter heating season in 2018, including the water-soluble ions (WSIIs), metallic

18 elements, and bacterial and fungal aerosols, were investigated. Terrestrial air masses

19 constituted a larger proportion of 59.94%, particularly during severe air pollution

20 episodes (up to 90%), exhibiting higher concentrations of $PM_{2.5}$ (240 μg/m^3) and

21 carrying more water-soluble ions and metal elements. A relative shift towards marine

22 air-mass with respect to pollution elimination stage was observed. The terrestrial air

23 mass harbors more animal parasites or symbionts, and human pathogens from

24 anthropogenic emission, such as *Deinococcus*, *Sphingomonas*, *Lactobacillus*,

25 *Cladosporium* and *Malassezia*. In comparison, saprophytic bacteria and fungi, such as

26   hydrocarbon degradation and gut bacteria from *Comamonas*, *Streptococcus*,

27   *Novosphingobium*, and *Aerococcus*, saprophytic *Aspergillus*, were the most prevalent

28   species in marine air mass. Mixed air-mass revealed the intermixing processes of

29   terrestrial and marine sources. This is a consequence of the amalgamation of

30   microorganisms from both terrestrial soils, animals, plants, and marine environments

31   during transportation. Correlation analysis suggested a higher correlation between

32   microorganisms and continental air mass, such as K^+, Mg^{2+}, and Ca^{2+} from soil dust.

33   Present study on constituents and amalgamation of bioaerosols over coastal atmosphere

34   encompassing distinct airmasses presume critical importance in comprehending the

35   terrestrial and marine air mass transport, intermixing processes and health implications.

36   **Keywords:** $PM_{2.5}$, water-soluble ions, terrestrial air mass, marine air mass, bioaerosols

1. Introduction

37

38   Bioaerosols, encompassing bacteria, fungi, viruses, pollen, cellular debris, etc., are

39   crucial aerosol particles in the atmosphere. These particles, ranging in size from 0.1 to

40   100 μm, are abundant in microorganisms or biomolecules that demonstrate life activity

41   (Górny, 2020; Urbano et al., 2011; Vishwakarma et al., 2023). Notably, bacterial

42   aerosols which can be either free-floating or attached to airborne particles, typically

43   measure between 0.3 and 10 μm (Tamer Vestlund et al., 2014). Such characteristics

44   allow for extended residence times and a heightened likelihood of long-distance

45   transport from local to distant regions within the atmospheric circulation (Galbán et al.,

46   2021; Smets et al., 2016). Fungal aerosols are prevalent in Earth's near-surface

47   atmosphere, with their origins being diversely found in water, soil, and plants (Eduard,

48   2008). Furthermore, the presence of pathogenic bacteria and fungi in the atmosphere

49   may lead to significant penetration efficiency within the human respiratory system

50   (Fakunle et al., 2021; Jiang et al., 2022; Smets et al., 2016).

51   Relevant studies have demonstrated that the concentration and community structure

52   of bacterial and fungal aerosols are significantly influenced by air emission sources,

53   atmospheric circulation, meteorological conditions, geographical and topographical

54   factors (Li et al., 2019; Liu et al., 2021; Núñez et al., 2019; Núñez et al., 2021). The

55   geographical and topographical factors, such as terrestrial and marine environments

56   exhibit significant differences in bioaerosol sources and pollution characteristics.

57   Studies focusing on continental environments, particularly in large inland cities like

58   Beijing (Zhang et al., 2019; Zhang et al., 2022), Xi'an (Wang et al., 2020; Yao et al.,

59   2019), Jinan (Wei et al., 2020) , and Nanjing (Hu et al., 2020) during severe air pollution,

60   have revealed that continental pollutant emissions significantly contribute to air

61   pollution. Furthermore, transboundary transportation of terrestrial air masses on hazy

62 days plays a crucial role in the progression of regional heavy pollution (Xie et al., 2020).

63 The ocean serves as both a potential source and sink for airborne microorganisms

64 (Archer et al., 2020; Mayol et al., 2017), exhibiting the intricate interplay between

65 continental communities and their transmission across oceanic regions (Bhangar et al.,

66 2015; Cho and Hwang, 2011; DeLeon-Rodriguez et al., 2013). Bioaerosols from the

67 oceans maybe influenced by long-distance transport from continental sources, such

68 as plants and human pathogens (Elbert et al., 2007; Sharoni et al., 2015). Studies have

69 shown that the concentration and diversity of bacterial and fungal aerosols from marine

70 are typically lower than those derived from continental sources (Cao et al., 2024; Shi et

71 al., 2022; Xue et al., 2022). Xu et al. (2019) undertook a thorough investigation of

72 bacterial abundance in Mt. Tai, China. Their findings indicate that variations in airmass

73 from diverse sources could potentially influence the chemical composition of $PM_{2.5}$.

74 This in turn prompts shifts in bacterial groups. Limited

75 studies have examined the impacts of terrestrial and marine air masses on chemical

76 constituents and microbial aerosols (Aswini and Hegde, 2021; Lang-Yona et al., 2022;

77 Shi et al., 2022). Generally, the chemical aerosols can be affected by various sources of

78 air masses, which may include local aerosols or remotely transported aerosols. However,

79 it remains unclear whether different air trajectories contribute to the formation of

80 bacterial communities within these particles. There is a dearth of research focused on

81 determining the chemical and biological composition of coastal cities affected by these

82 air masses.

83   Coastal atmospheres frequently display a complex amalgamation of terrestrial and

84 marine aerosols, with their characteristics being markedly influenced by the origins of

85 the air masses. Therefore, the costal aerosols provide the ideal conditions for

86 understanding the mixing processes taking place between natural and anthropogenic air

87   masses from terrestrial and marine, respectively. Meteorological factors as well as

88   changes in pollutant concentrations and compositions exhibit significant variability

89   under different air masses. The average abundance of bacterial and fungal spores in the

90   atmospheric boundary layer over land is approximately 1.9×10^4 cells/m^3 and 2.4×10^4

91   cells/m^3, respectively (Mayol et al., 2014; Spracklen and Heald, 2014). Marine

92   microbial aerosols can be released from ocean micro-surface or transported from land

93   (Prospero et al., 2005) and settle thousands of kilometers away from their source of

94   release (Mayol et al., 2014), exerting significant impacts on ecological and climate

95   systems. Atmospheric aerosols traversing marine regions have been documented to

96   contain marine bacteria from the *Cyanobacterial* and *α-Proteobacterial* classes, which

97   are predominantly observed in air samples (Aller et al., 2005). Gong et al (2020a)

98   conducted an examination of the microbial composition along Qingdao's coastlines,

99   revealing a higher proportion of bacteria to total microorganisms in samples from

100   continental sources compared to marine sources. Xu et al (2020b) studied the diversity

101   of bacterial populations in PM$_{2.5}$ across urban and rural areas of Shanghai, finding that

102   airborne microbial communities over coastal cities are more influenced by long-

103   distance transport than those inland. Some marine bacteria persist in aerosols after land

104   transportation (Raman and Wagner, 2011). Air samples taken at high altitude zones in

105   coastal Europe and Japan demonstrated the continental transportation of marine

106   microorganisms (Maki et al., 2014). The introduction of marine bacteria into

107   tropospheric free space may modify the airborne microbial composition in continental

108   regions (Amato et al., 2007; Cáliz et al., 2018; Cho and Hwang, 2011; Maki et al., 2014;

109   Polymenakou et al., 2008).

110   Weihai, a coastal city situated at the confluence of the eastern Asia continent and the

111   Northwest Pacific Ocean, is prone to the impact of marine and terrestrial air masses.

112   Typically, Weihai maintains low pollutant emissions with an annual average $PM_{2.5}$

113   concentrations below 35 μg /m^3 throughout the year. However, during winter and spring

114   periods, regional air pollution intensifies, leading to severe air quality issues. This is

115   due to increased inter-regional transportation, resulting in daily average $PM_{2.5}$

116   concentrations greater than 150 μg/m^3 (Wei et al., 2020). In this study, we conducted an

117   integrated atmospheric observation experiment to examine the potential impact of

118   terrestrial and marine air-mass on the constituents and amalgamation of bioaerosols

119   over the coastal atmosphere. This approach allows for a comprehensive exploration of

120   the effects of sea-land air mass exchange on the spatial and temporal distribution of

121   aerosols, as well as potential intermixing processes in coastal regions.

122   **2. Materials and methods**

123   **2.1 Sample collection**

124   The sampling site was situated at the national air sampling station of Shandong

125   University (37.53°N, 122.06°E), approximately 1-2 km from the coast (Fig. S1). The

126   sampling platform was mounted on the rooftop of a building, positioned about 15 m

127   above ground level, and devoid of any significant obstructions. $PM_{2.5}$ samples were

128   gathered between January to March, 2018, during the winter heating season in

129   northern China. Two parallel $PM_{2.5}$ sampler with a particle size of 2.5 ± 0.2 μm were

130   utilized equipped with 47 mm Quartz membrane for the collection and analysis

131   of atmospheric $PM_{2.5}$, inorganic ion, metal elements, and microbes. Prior to use, the

132   Quartz membranes were cauterized in a muffle furnace at 450°C for 6 hours to eliminate

133   carbonaceous and contaminant materials. Samples were collected twice a day

134   (07:00~19:00; 19:00~07:00 the following day). Ultimately, 300 $PM_{2.5}$ samples were

135   obtained, out of which 40 samples were chosen for subsequent analysis based on

136   air mass categories. Blank samples were obtained using membranes without sampling,

137   and all these samples were stored at -80°C until further analysis.

138   Meteorological parameters, such as air temperature, relative humidity, wind direction,

139   and wind speed, were monitored in situ utilizing a PC-4 automatic weather station (PC-

140   4, JZYG, China). The hourly concentrations of $PM_{2.5}$, PM_{10}, CO, SO_2, NO_2, and O_3

141   were systematically retrieved from the National Ambient Air Quality Monitoring

142   System (http://www.cnemc.cn/).

143   The mass concentration of $PM_{2.5}$, water-soluble ions, and metal elements were

144   quantified post sampling. The membranes were meticulously weighed utilizing a

145   Mettler XP-6 balance with an accuracy of 10^{-6} g. Prior to the weighing, the membranes

146   were maintained in a controlled environment with consistent temperature and humidity

147   for a duration of 24 hours. Ion chromatography (ICS-2100, Chameleon 6. 8, AS-DV

148   autosampler Thermo Fisher) was employed to ascertain the presence of water-soluble

149   ions such as Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, NO_3^-, SO_4^{2-}, and NH_4^+. These ions were extracted

150   via ionized water ultrasonication, subsequently separated through anion or cation

151   column exchange, and identified using a conductivity detector. Metallic elementals

152   were scrutinized using microwave digestion extraction (ETHOS ONE, Milestone), with

153   the concentrations determined by ICP-MS or ICP-OES (Thermo Fisher). This analysis

154   encompassed elements as Al, Fe, Ti, Mn, Co, Ni, Cu, Zn, Ga, Sr, Cd, Sn, Sb, Pb, V, Cr,

155   and As.

156   **2.2 Air mass clustering and classification**

157   The potential sources and transport pathways of air mass were examined using the

158   MeteoInfo   backward   trajectory   model   (MeteoInfo   3.7.4   –   Java,

159   http://www.meteothink.org/downloads/index.html) developed by the Chinese Academy

160 of Meteorological Sciences. For each sample, backward trajectories were simulated one-

161 hour intervals and estimated over a 24-hour period. The meteorological data were

162 sourced   from   GDAS1   (ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1/).

163 Backward trajectories of air masses at an altitude of 500 meters were categorized and

164 clustered, with daily plots illustrating these trajectories. In this study, we defined a

165 marine air-mass sample if more than 90% of the masses originated from the ocean;

166 terrestrial air-mass sample if more than 90% originated from the continent; and mixed

167 if the proportions of terrestrial and marine air masses were similar or accounted for

168 more than 40% of the total air masses.

169 **2.3 DNA extraction and qPCR amplification**

170   Microbial genomic DNA in $PM_{2.5}$ samples were procured from filters utilizing the

171 Fast-DNA^{TM} SPIN kit for soil (MoBio Laboratories, Carlsbad, CA, USA). The

172 extracted DNA was measured via a Nanodrop spectrometer (Nanodrop 2000, Thermo

173 Scientific USA) to ascertain the concentration. The quantitative polymerase chain

174 reaction (qPCR) was employed to identify bacterial 16S rRNA and fungal ITS gene

175 copy numbers in $PM_{2.5}$, as well as to estimate the count of bacteria and fungi per cubic

176 meter of air. The bacterial 16S V3-V4 variable region was selected for PCR

177 amplification using primer 338F (5'-ATCTACGGGGGGCAGCAG-3') and 806R

178 (5'GGACTACHVGGGTWTCTAAT-3') (Masoud et al., 2011). The fungal ITS region

179 were amplified using the primers ITS1 (5′-CTTGGTCATTTAGAGGAAGTAA-3′) and

180 ITS2 (5′-GCTGCGTTCTTCATCGATGC-3′) (Liu et al., 2021).

181   The PCR amplification conditions comprised an initial denaturation at 95°C for

182 duration of 5 minutes, succeeded by 30 seconds at 95°C, 30 seconds at 50°C, and then

183 35 cycles at 72°C for 40 seconds, followed by a final extension phase at 72°C

184   for seven minutes to ensure comprehensive amplification. Fluorescent signals were

185   gathered during this extension phase. For each sample, qPCR was conducted in

186   triplicate, with ultrapure water serving as a negative control to identify potential PCR

187   contamination. Standard curves were constructed using *E. coli* harboring the 16S rRNA

188   gene and *Streptomyces* plasmids containing the ITS gene. Additionally, gradient

189   dilutions of these plasmids were performed, ranging from 10^2-10^7 copies/µL. The FTC-

190   3000 real-time quantitative PCR system was employed for standard curve construction

191   and data processing.

192   **2.4 16S rRNA and ITS gene sequencing and data processing**

193   Similarly, the V3-V4 region of the bacterial 16S rRNA and the fungal ITS1 gene

194   were targeted for PCR amplification utilizing barcode-specific primers 338F-806R and

195   ITS1F-ITS2, respectively. To ensure optimal amplification efficiency and precision,

196   a high-fidelity enzyme (Phusion® High Fidelity PCR Master Mix from NewEngland

197   Biolabs) in conjunction with GC buffer was employed during PCR amplification.

198   This procedure entailed a pre-denaturation step at 98°C for one minute, followed by 30

199   cycles of 98°C for ten seconds; 58°C for bacteria (56°C for fungi) for 30

200   seconds; and 72°C for an additional 30 seconds. This cycle was repeated seven times,

201   concluding with a final extension at 72°C for five minutes, after which the samples

202   were stored at 4°C. Following amplification, the samples were purified using the

203   Agencourt Ampure XP kit (from Beckman Coulter, Brea, CA, USA). Subsequently,

204   they were combined to achieve equimolar concentrations and analyzed on the Illumina

205   MiSeq platform provided by Illumina, Inc. in San Diego, CA.

206   Following sequencing, the barcode sequences of each sample were extracted and

207   subsequently stored in fastq format utilizing the QIIME toolkit (Caporaso et al.,

208   2010). Sequences shorter than 200 base pairs, with mass of less than 25, and containing

209   ambiguous bases underwent quality control screening using Trimmomatic (Bolger et

210   al., 2014) and Mothur (Schloss et al., 2009). The assembled sequences were then de-

211   duplicated and trimmed to equal length. De-chimerized sequences were eliminated

212   (Edgar, 2013), and sequences were clustered into operational taxonomic units (OTUs)

213   using UPARSE at a 97% identity threshold. Subsequently, individual OTUs were

214   removed. Taxonomic assignments were determined using a Basic Local Comparison

215   Search Tool (BLAST) search against the bacterial SILVA Archived SSU Ref NR 99 132

216   dataset and fungal UNITE ITS reference v7.2 dataset. The original raw sequences were

217   deposited in the Sequence Read Archive under the accession number PRJNA1096829.

218   **2.5 Statistical analysis**

219   The bacterial community functional analysis was conducted using FAPROTAX, a

220   manually constructed database that maps prokaryotic taxa to metabolic or other

221   ecologically functions, such as sulfur, nitrogen, hydrogen, and carbon cycling.

222   FUNGuild (Fungi Functional Guild) facilitates the taxonomic analysis of fungal

223   communities by employing microecological guilds. Samples from different air-mass

224   samples were examined for intergroup species variability, based on community

225   abundance data. This was achieved using rigorous statistical methods to identify species

226   demonstrating differences in abundance within the microbial communities of different

227   groups, and hypothesis testing to evaluate the significance of these observed differences.

228   Statistical analysis, including Analysis of variance (ANOVA) and Kruskal Wallis tests,

229   were employed to discern bacteria with varying abundances between samples and

230   groups. A p value less than 0.05 was considered significant. ANOVA was used to

231   analyze variation in a response variable measured under conditions defined by discrete

232 factors (Martin G, 2008). The Kruskal-Wallis test determines whether there is a

233 statistically significant difference between the medians of three or more independent

234 groups (Kassambara, 2019). The disparities and primary influencing factors on

235 microbial community between terrestrial and marine air masses were assessed using the

236 Mantel correlation analysis, a method predominantly utilized in ecology

237 to examine the relationship between community and environmental variables. The

238 statistical was determined using the Spearman's rank correlation coefficient, with

239 significance levels set at $p<0.05$ and $p<0.01$.

240 **3. Results and Discussion**

241 **3.1 Air mass backward trajectory**

242   The MeteoInfo backward trajectory model was employed to simulate the trajectories

243 of air masses at an altitude of 500 m in Weihai over 24-hour period from January to

244 March 2018, identifying and classifying potential sources of air mass transport (Fig.

245 S1). The terrestrial air mass accounted for 59.94%, exhibiting an average $PM_{2.5}$

246 concentration of 36.15 ± 26.52 μg/m^3. Severe air pollution episodes occurred on January

247 20, March 19, and March 24, with $PM_{2.5}$ concentration reaching 240 μg/m^3, 153 μg/m^3,

248 and 119 μg/m^3, respectively. During reginal air pollution, the terrestrial air masses

249 primarily influenced Weihai were typically originating from the Beijing-Tianjin-Hebei

250 region and the surrounding areas. The significant ownership of motor vehicles is

251 identified as a major source of nitrogen oxides (Yang et al., 2018). Furthermore, the

252 primary contributors to emissions in the terrestrial air mass of this region are the dense

253 population, industrial and agricultural activities (Wei et al., 2019).

254   The marine air masses mainly originate from the Yellow Sea, traversing the marginal

255 sea of the western Pacific Ocean before making contact with the land prior to reaching

256   the study area. A total of 14% of the sampling days were influenced by these marine air

257   masses, resulting in an average $PM_{2.5}$ concentration of 23.99±11.00 μg/m³. Mixed air

258   masses, characterized by simultaneous influence from the northwestern winds of Inner

259   Mongolia and the offshore air masses of the Yellow Sea, accounted for 27% of all

260   affected sampling days. These mixed air masses yielded an average $PM_{2.5}$

261   concentration of 45.11±12.69 μg/m³. In comparison, under the influence of mixed air

262   masses, pollutant concentrations were notably high. During the spring dust season,

263   there is a notable increase in the proportion of mixed air mass. This elevated

264   concentration of particulate matter correlates with the transmission of sand dust (Xie et

265   al., 2020).

266   Three heavy pollution episodes were examined to investigate air mass shifts during

267   pollution (Fig. 1, Fig. S2). Generally, the initiation and development stages of pollution

268   events were predominantly characterized by terrestrial and mixed air masses.

269   Conversely, during the stage of pollution mitigation, marine air masses were the

270   primary contributors. For instance, the pollution episode I occurred from January 19 to

271   January 21. During this period, the western terrestrial air mass was predominantly

272   responsible for initiating the pollution, evidenced by a $PM_{2.5}$ concentration of 51.35

273   μg/m³. As the pollution progressed, it transitioned into a mixed air mass with a $PM_{2.5}$

274   concentration peaking at 240 μg /m³. This change was accompanied by significant

275   increases in WSIIs and elemental concentrations. The introduction of marine air mass

276   from the west led to the elimination of the pollution, resulting in an average $PM_{2.5}$

277   concentration reduction to 7.92 μg /m³. This reduction was also associated with lower

278   WSIIs and elemental concentrations, due to the scavenging effect of the marine air mass.
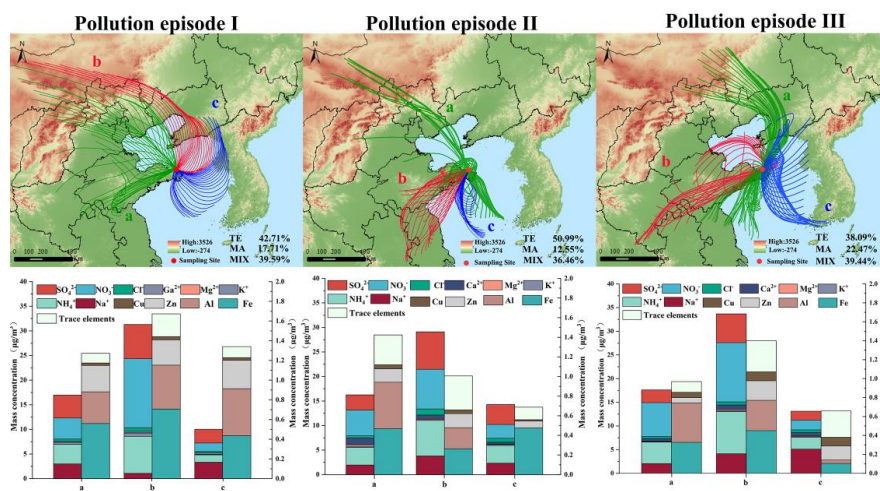
Fig. 1 Transformation of terrestrial and marine air masses of three severe air pollution episodes. A, the pollution initiation; b, the pollution development; c, the pollution elimination. TE, terrestrial air mass; MA, marine air mass; MIX, mix air mass.

3.2 Water-soluble ions and metal elements concentration

During the sampling period, the daily average concentration of water-soluble ions in $PM_{2.5}$ was 22.92 ± 12.19 μg/m^3, exhibiting a range of variation from 4.58 to 78.14 μg/m^3. Notably, NO_3^- had the highest concentration (26.94%, ranging from 6.4% to 52.6%), succeeded by SO_4^{2-} (21.94%, between 9.4% and 33.4%) and NH_4^+ (20.26%, from 5.8% to 35.6%). The concentrations of SO_4^{2-}, NO_3^- and NH_4^+, constituted the majority of total ion concentration in other studies conducted in Beijing, Shanghai, and Guangzhou (Hu et al., 2014; Pathak et al., 2008; Zhou et al., 2012). The concentrations of NO_3^-, SO_4^{2-}, and NH_4^+ were significantly influenced by both terrestrial and mixed air masses, with the latter exhibiting a more pronounced effect (NO_3^-, 10.65 ± 3.26 μg/m^3; NH_4^+, 7.39 ± 3.30 μg/m^3; SO_4^{2-} 6.76 ± 1.77 μg/m^3) (Fig. S3). In marine air-mass samples, a notably lower concentration of water-soluble ions was observed, with the concentration of 13.01 ± 7.43 μg/m^3, 27.94 ± 13.61 μg/m^3 and 30.38 ± 11.38 μg/m^3 in marine, terrestrial and mixed air masses, respectively.

297   A high concentration of Na^+ was observed, with a range that from 3.15 ± 1.69 µg/m^3,

298   which accounted for 14.47% of the total water-soluble ion. The concentrations of Na^+

299   and Mg^{2+} did not significantly differ in the three types of air-mass samples. These two

300   ions are typical components of sea salt. The average ratio of Mg^{2+}/Na^+ was found to be

301   0.11, which closely aligns with the value of 0.12 in seawater. This suggests potential

302   sources from marine environments (Sun et al., 2022). The concentrations of K^+

303   0.24±0.20 µg/m^3 and 0.26±0.10 µg/m^3 in the terrestrial and mixed air-mass samples,

304   and was twice as high as those in the marine air-mass samples (0.11±0.05 µg/m^3), which

305   suggests an important contribution from anthropogenic emissions. The concentrations

306   of Cl^-, and Ca^{2+} in both terrestrial and marine air mass samples exhibited similarity.

307   However, a marked increase was observed in mixed air masses. Cl^- mainly comes from

308   sea salt, coal and biomass combustion, and Ca^{2+} is mainly affected by sand dust in

309   spring (Liang et al., 2021). The coastal city was more affected by sea salt, and the coal-

310   fired power plants in winter heating season. The high concentration in mixed air mass

311   were associated with the sea salt, coal combustion, dust events, and construction

312   activities (Sun et al., 2021).

313   The concentration of various elements in different air-mass samples are depicted in

314   Fig. S3. The top ten elements identified in PM$_{2.5}$ were found to be Fe, Al, Zn, Cu, Sn,

315   Pb, Mn, Ti, Ni, and V, in descending order. The metal elements were categorized into

316   macro and trace elements. The macro metals, specifically Iron (Fe), Aluminum (Al),

317   and Zinc (Zn), constituted a significant proportion of the total heavy metal elements,

318   accounting for 34%, 25%, and 23% respectively. In general, the concentration of both

319   macro and trace metal elements in marine air masses was found to be lower than that in

320   terrestrial and mixed air masses. The V/Ni ratio serves as an indicator of ship emission

321   influence (Celo et al., 2015; Viana et al., 2009). A ratio of V/Ni exceeding 0.7 suggests

322   a substantial impact from ship emission sources, typically used as an indicator of ship

323   emission influence in coastal cities, in conjunction with the trajectories of air mass

324   transport. (Zhang et al., 2014). The V/Ni ratio in marine air-mass samples was found to

325   be 0.78, a value significantly higher than that of both terrestrial and mixed air masses.

326   **3.3 Microbial community over coastal atmosphere**

327   Airborne bacterial and fungal concentration in $PM_{2.5}$ were $1.99\pm1.46\times10^5$ cells/m^3

328   and $3.39\pm1.10\times10^4$ cells/m^3, respectively. A high concentration was observed in

329   terrestrial air-mass samples, with the average value of $4.72\pm3.93\times10^5$ cells/m^3 and

330     $6.37\pm1.70\times10^4$ cells/m^3 for bacteria and fungi, respectively. The terrestrial air masses

331   came from the inland areas, which carried more microorganisms from anthropogenic

332   activities and natural sources with high resistant to high temperatures, dryness, and

333   strong ultraviolet rays (Gong et al., 2020). Microbial concentration in marine air-mass

334   samples was significantly lower, with the average concentrations of bacteria and fungi

335   being $4.91\pm1.82\times10^4$ cells/m^3 and $6.15\pm3.09\times10^3$ cells/m^3.
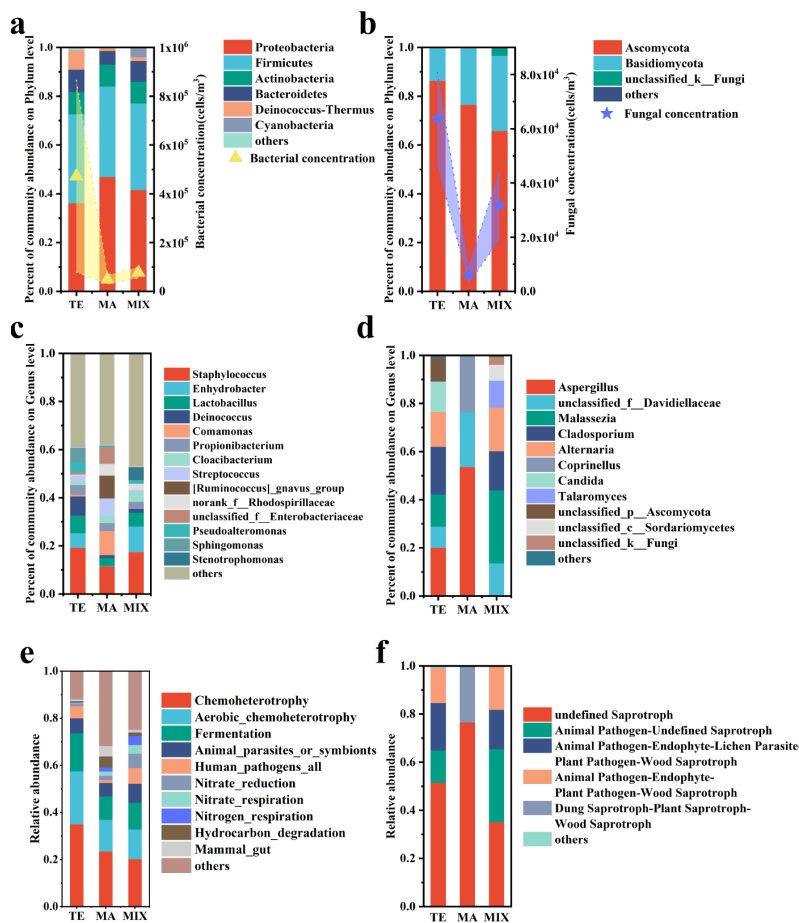
Fig. 2. Bacterial and fungal species and function analysis influenced by different air masses. Bacterial and fungal concentration, main phylum (a), (b), Bacterial and fungal genus (c), (d), and bacterial and fungal community function (e), (f) are indicated.

The microbial community structure exhibited significant variation influenced by marine and terrestrial air masses (Fig. 2). Predominantly, *Proteobacteria* (40.06%), *Firmicutes* (36.30%), *Actionbacteria* (8.97%), *Bacteroidota* (8.29%), and *Deinococcus-Thermus* (4.59%) were identified as the most abundant bacteria. Notably, *Actionbacteria* and *Deinococcus-Thermus* were found in high concentrations in samples from terrestrial air masses. In particular, *Deinococcus-Thermus* demonstrated a relative abundance of 7.9% in terrestrial air mass samples, significantly surpassing

347   that of marine (1.2%) and mixed air mass samples (1.5%). As a radiation-resistant

348   bacterium, *Deinococcus-Thermus* is capable of withstanding harsh environmental

349   conditions (Callegan et al., 2008; Rainey et al., 2007). The majority of these bacteria

350   possess the ability to produce spores or spore germs. These spores are capable of

351   withstanding harsh conditions such as low temperatures, aridity, and radiation during

352   long-distance transmission processes, thereby ensuring their survival throughout this

353   process (Griffin, 2003). *Cyanobacteria* exhibit a higher concentration in both terrestrial

354   and mixed air-mass samples. As typical soil bacteria found in desert environments, they

355   are prone to forming soil crusts in arid regions (Li et al., 2014; Li et al., 2013).

356   *Proteobacteria* was the most prevalent taxon in marine air-mass samples. The

357   predominant metabolic activity in deep-sea sediments is attributed to this group, with

358   major taxa being found in marine sediments (Huang et al., 2021).

359   The predominant bacterial genera included *Staphylococcus* (21.02%),

360   *Enhydrobacter* (6.43%), *Lactobacillus* (6.03%), *Deinococcus* (4.56%), and

361   *Propionibacterium* (3.48%). Terrestrial air masses and mixed air masses had similar

362   bacterial community composition. The abundance of bacteria such as *Staphylococcus*,

363   *Enhydrobacter*, *Lactobacillus*, *Deinococcus* is higher influenced by terrestrial and

364   mixed air masses. *Staphylococcus* is a pathogenic bacterium widely found in human

365   skin, nasal cavity, throat, and intestines (Cheung et al., 2021). Relative abundance of

366   bacteria such as *Comamonas*, *Streptococcus*, *Ruminococcus*, and *Enterobacteriaceae*

367   were higher in marine air-mass samples. *Comamonas* is a gram-negative bacillus,

368   inhabiting in natural soil, freshwater, and animal gut. They have also been isolated from

369   industrial environments, such as activated sludge and contaminated soils, as well as

370   from hospital environments and clinical samples. *Comamonas* is associated with

371   environmental bioremediation, and is considered an important environmental bacterium

372   rather than a human pathogen (Ryan et al., 2022). *Streptococcus* are mostly found in

373   the oral and gastrointestinal tracts of a variety of mammals and have not been shown to

374   play a role in human infections to date (James et al., 2015). *Ruminococcus*, and

375   *unclassified Enterobacteriaceae* are gut microorganisms that may be related to the

376   marine fish and other animal gut microbes. Most bacteria carried by marine air masses

377   with high abundance are saprophytic in nature.

378   The dominant fungal phyla were *Ascomycota* (77.29%) and *Basidiomycota* (21.58%),

379   which were similar to the previously studies (Du et al., 2018; Liu et al., 2019; Zeng et

380   al., 2019). Fungal community influenced by terrestrial and mixed air masses were quite

381   similar, with relatively higher abundances of opportunistic pathogens such as

382   *Malassezia*, *Alternaria*, *Cladosporium*. In contrast the saprophytic *Aspergillus*,

383   *Davidiellaceae* and *Coprinellus* were abundant in marine air-mass samples.

384   **3.4 Community disparities influenced by terrestrial and marine air masses**

385   The community disparities influenced by terrestrial, marine, and mixed air masses

386   was conducted in Table S2, Table S3, Fig. 3 and Fig. 4. The bacterial enrichments

387   observed in the samples from terrestrial and mixed air-masses included *Deinococcus*,

388   *Lactobacillus*, and *Sphingomonas*. *Deinococcus* is known to tolerate high radiation and

389   adapted to harsh or extreme environments (Wei et al., 2020). *Lactobacillus*, a genus of

390   Gram-positive bacteria, has also been identified as abundant in atmospheric dust

391   (Federici et al., 2018; Xu et al., 2017). *Sphingomonas*, typically found in water bodies,

392   soil, and roots, can thrive in extreme environments (Hu et al., 2007; Sun et al., 2018).

393   It is usually more abundant in deserts and can be transported over long distances via air

394   masses. In contrast, *Comamonas* was identified as an indicator bacterium in marine air-

395   mass samples, which is dominant in coastal cities (Wei et al., 2020) and originates from

396   soil, activated sludge, and water (Yan et al., 2012).

397   For fungal community, *Aspergillus* (p=0.014) and *Malassezia* (p=0.041) were

398   significantly differentiated in different air masses. *Aspergillus* was 53.7% and 20.1% in

399   marine and terrestrial air-mass samples, respectively. *Aspergillus* is a dominant fungus

400   in offshore areas such as Qingdao, China (Li et al., 2011). Moreover, the Saprophytic

401   *Aspergillus* was also prevalent in clean samples during haze pollution episode and was

402   commonly detected on non-Haze days (Yan et al., 2016). Prior research has established

403   that *Aspergillus* is ubiquitously found in nature and non-polluted environments (Li and

404   Kendrick, 1995). *Malassezia* was higher in terrestrial and mixed air-mass samples,

405   which has been found to be widespread in a variety of animals. As a parasitic fungus,

406   *Malassezia* causes the majority of skin diseases, such as dandruff and seborrheic

407   dermatitis caused by *Malassezia sphericalis* (DeAngelis et al., 2007). The

408   *Cladosporium* is significantly higher in terrestrial and mixed air-mass samples than

409   marine air-mass samples, which is ubiquitous worldwide, commonly found in a wide

410   variety of plants and is frequently isolated from soils, paints, textiles, foodstuffs and

411   organic matters (Bensch et al., 2012; Ellis, 1977), known to be a common endophyte as

412   well as a foliar fungus (El-Morsy, 2000; Islam and Hasin, 2000). *Cladosporium* have

413   been commonly observed in terrestrial atmospheric environments, demonstrating the

414   potential origins from continental environments (Frączek et al., 2017; Han et al., 2019).
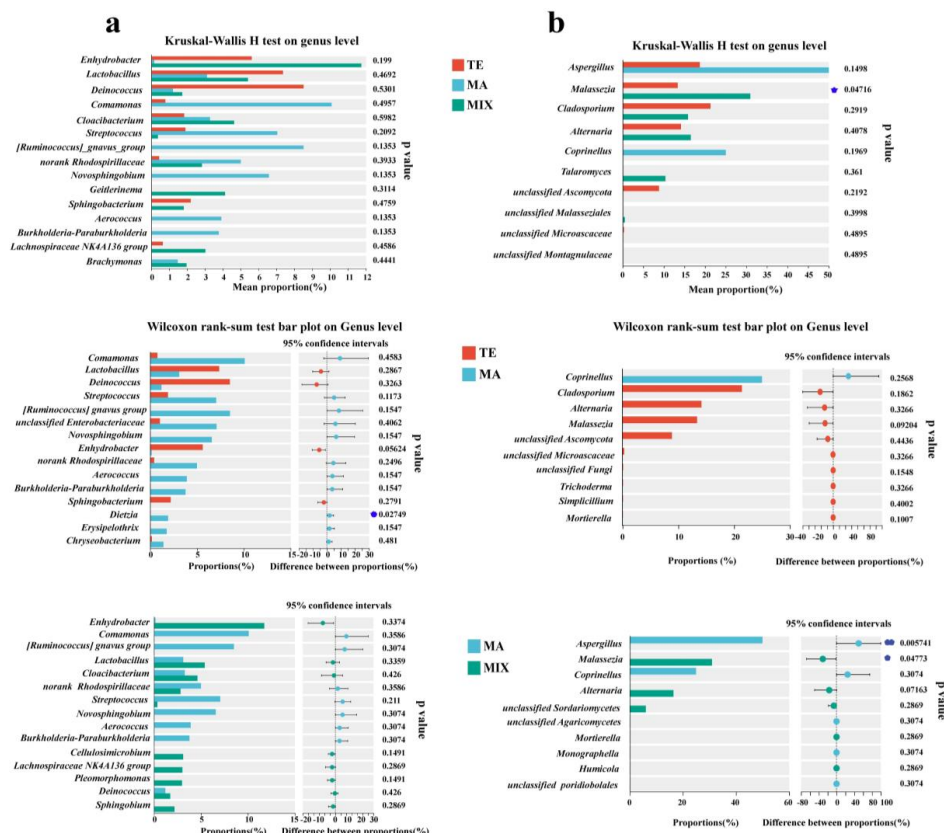
Fig. 3 Bacterial (a) and fungal (b) community composition disparities influenced by different air masses

The FAPROTAX analysis revealed that the primary bacterial ecological functions were chemoheterotrophy, aerobic chemoheterotrophy, fermentation, and human pathogens. These accounted for 74%, 47%, and 44% respectively in terrestrial, marine, and mixed air-mass samples. Notably, human pathogens and animal parasites or symbionts were more prevalent in terrestrial and mixed air-mass samples than in marine air-mass samples. Conversely, marine air-mass samples were enriched with mammal gut bacteria, as well as hydrocarbon and automatic compound degradation bacteria. Overall, the dominant airborne bacteria in the coastal city during winter primarily inhabited anthropogenic environments such as soil, water, and terrestrial ecosystems. Additionally, marine ecosystems served as a significant source of airborne microbes (Griffin, 2003; Xu et al., 2019).

The fungal community function in terrestrial and mixed air-mass samples were

430 similar, with undefined Saprotroph, Animal Pathogen-Undefined Saprotroph, Animal

431 Pathogen-Endophyte-Lichen Parasite, Animal Pathogen-Endophyte-Plant Pathogen

432 were the main functions, which totaled 99.27%, 99.98%, 99.27% in the terrestrial,

433 marine, and mixed air-mass samples. In particular, the prevalence of Saprotroph fungi

434 was observed higher in samples from marine air masses, such as those containing

435 *Aspergillus* (Xu et al., 2017). Notably, the fungi associated with terrestrial air masses

436 predominantly carried animal pathogens and exhibited greater pathogenicity, including

437 species such as *Malassezia* and *Alternaria* (Gandolfi et al., 2013; Masiol et al., 2012).
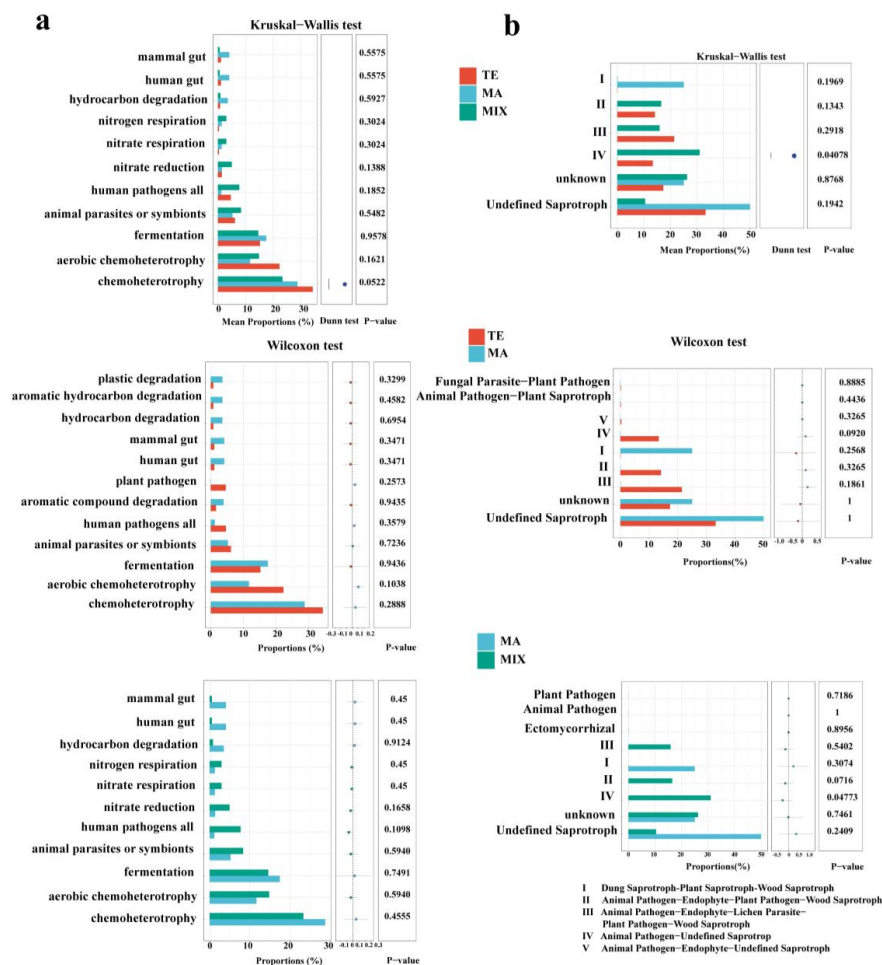
Fig. 4. Bacterial (a) and fungal (b) community function disparities influenced by different air masses

3.5 Implications of environmental factors on coastal airborne microbes

441   To understand the contribution of marine and terrestrial air masses to microbial

442   aerosols in the coastal city, the Mantel correlation analysis between microbial aerosols

443   and environmental factors were performed (Fig.5, Fig.S4). Influenced by terrestrial and

444   mixed air masses, bacterial community was significantly positively correlated with

445   SO_4^{2-} ($P<0.05$), and bacterial and fungal communities were positively correlated with

446   continental ions such as K^+, Mg^{2+}, and Ca^{2+} (Shi et al., 2022). Moreover, bacterial and

447   fungal concertation was positively correlated with NO_2 ($P < 0.05$) and significantly

448   negatively correlated with PM_{10} (P<0.01).

449   Air parcels transported over long distances appear to harbor diverse microbial

450   populations (DeLeon-Rodriguez et al., 2013; Kakikawa et al., 2009). The long-distance

451   transportation of dust particles from the northwestern winds in the Inner Mongolia

452   region may have changed the community structure and abundance (Castañer et al., 2017;

453   Squizzato and Masiol, 2015). Dust-borne bacteria (*Staphylococcus*, *Delftia*,

454   *Pseudoalteromonas* and *Deinococcus*) were injected into the atmosphere during dust

455   events, and most of them accompanied the dust transportation to the downwind of Asian

456   Dust including the coastal city of Weihai. Influenced by the mixed air masses, bacterial

457   community was significantly positively correlated with K^+ ($P<0.01$) and negatively

458   correlated with PM_{10} ($P<0.05$). Similarly, microbial communities showed high

459   positively correlated with ions from continental sources, such as K^+, Mg^{2+}, and Ca^{2+},

460   which indicated that microorganisms carried by mixed air masses were mostly from

461   continental sources (Bates et al., 1992). A negative correlation was observed between

462   microbial communities and wind directions influenced by terrestrial and mixed air

463   masses. Wind blowing from continent or marine may play important role in microbial

464   community diversity (Jones and Harrison, 2004). Moreover, influenced by mixed air

466   mass, temperature have a greater impact on fungal community, which was positively

467   correlated with *Malasseziales* and *Davidiellaceae*.

468   The marine air masses are generally clean and have a strong scavenging effect on air

469   pollutants. Notably, bacterial community had high correlation with sea salt ions such as

470   Mg^{2+}, and certain fungi species such as *Talaromyces*, *Monographella* and *Phoma* were

471   correlated with Na^+ (*P < 0.05*), which suggesting an obvious influencing form marine

472   sources. A positive correlation between air temperature and certain microorganisms

473   (*Aerococcus*, *Cloacibacterium*, *Sphingobium*, *Enhydrobacterium*, *Davidiellaceae*,

474   *Malasseziales*) indicates that the increase in air temperature in spring favors the survival

475   of airborne microbes, especially for pathogenic bacteria.
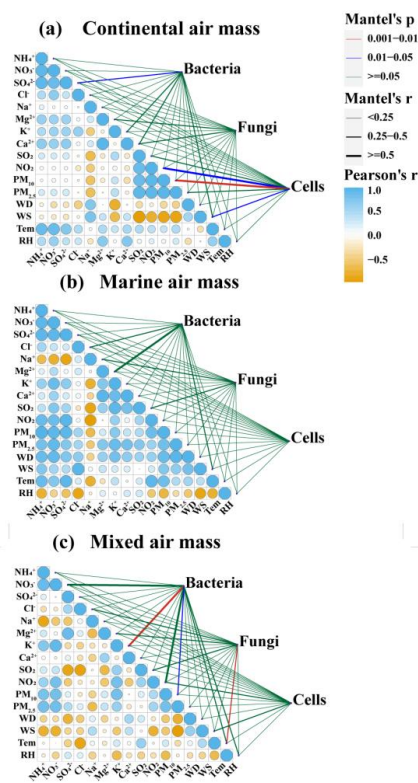
Fig.5 Relationships between microorganisms and environmental factors influenced by different air masses, terrestrial air masses (a) marine air masses (b) and mixed air masses (c)

This work shows that enormous levels of haze aerosols from the terrestrial and mixed air mass can be rapidly transported into the coastal city during regional haze pollution. During the long-distance transportation of air masses, a comprehensive mixture of soil-derived, biogenic, and anthropogenic microorganisms from terrestrial air masses, and aquatic, saprophytic, gut microorganisms from marine environments are fully mixed. This mixing is particularly evident in the samples of mixed air masses. Among that, not only large numbers of chemical components but also bacteria and fungi, as well as opportunistic pathogens, were transported into the coastal city. Microbial communities were strongly correlated with haze aerosols, e.g., WSIIs in $PM_{2.5}$ from terrestrial and

489   mixed air mass. The primary influence on terrestrial air mass was anthropogenic

490   emissions, with coal combustion for winter heating and biomass burning being the

491   predominant pollution sources. Moreover, dust events in spring carried higher

492   concentrations of particulate matter. These air pollutants can act as initial source of

493   bioaerosols such as bacteria and fungi, thereby providing a site for attachment

494   reproduction (Jiang et al., 2022). Additionally, water-soluble ions in PM, primarily

495   secondary ions, sulfate, nitrate, and ammonium ions, can supply essential nutrients for

496   microbial growth (Fan et al., 2019). This explanation elucidates the increased

497   concentration of particulate matter and microorganisms in terrestrial air mass during

498   heavy pollution. Simultaneously, it is pertinent to highlight that during pollution

499   incidents, the terrestrial air mass intensifies the pollution process. This intensification

500   results in a significant increase in the proportion of pathogenic microorganisms.

501   Conversely, the marine air mass facilitates the removal of pollution, introducing a

502   higher number of saprophytic microorganisms (Fig. 6). The presence of

503   microorganisms in marine air masses exhibited a significant correlation with sea salt

504   ions, specifically Na^+ and Mg^{2+}. These ions, when introduced into the atmosphere, form

505   aerosolized particulate matter. Additionally, meteorological factors exerted a more

506   pronounced influence on the concentrations of pollutants within marine air-mass

507   samples. Our research suggests that populations in coastal cities may also be susceptible

508   to exposure to these bioaerosols and pathogens, which are transported over long

509   distances during regional haze events.
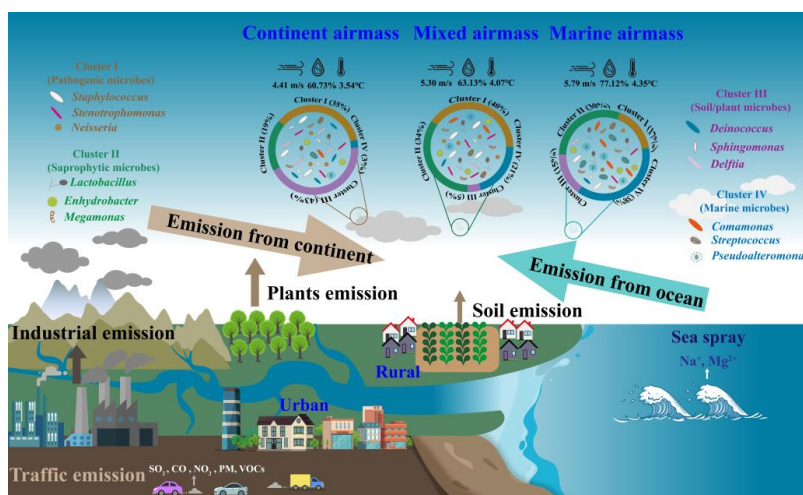
Fig. 6 Implications of sea-land differences and various environmental factors, such as air pollutants and meteorological conditions, on the composition of airborne microbial communities. This is influenced by a range of air mass transmissions, including terrestrial, marine, and mixed air masses.

4. Conclusion

This study evaluated the potential influence and the mixing effects taking place between natural and anthropogenic aerosols respectively from terrestrial and marine air-mass on biological constituents of PM$_{2.5}$ over coastal atmosphere. The concentration of water-soluble ions, metal elements and bioaerosols were higher in terrestrial and mixed air-mass samples than marine air-mass samples. However, the V/Ni ration greater than 0.7 in marine air-mass samples indicated significant influence from marine ship emissions. Bacterial and fungal community influenced by terrestrial and mixed air masses were enriched in animal pathogens, including the *Staphylococcu*, *Comamonas*, *Malassezia* and *Alternaria.* In contrast, marine air mass carried more saprophytic microbes, such as *Ruminococcus*, *Enterobacteriaceae*, *Aspergillus,* and *Davidiellaceae*. Potential implications of environmental factors on airborne microbes based on Mantel correlation analysis suggested that the bacterial community was mainly significantly

528 correlated with SO_4^{2-}, K^+, Mg^{2+}, and Ca^{2+} and PM_{10} influenced by terrestrial and mixed

529 air masses, which were mainly related to continental sources. In contrast, influenced by

530 marine air masses, bacterial and fungal community were strongly correlated with sea

531 salt ions, such as Na^+ and Mg^{2+}. The effects of meteorological factors on bioaerosols

532 vary significantly under different dominant air masses. The microbial communities

533 were found to be negatively correlated with wind direction, influenced by both

534 terrestrial and mixed air masses. Conversely, the predominant bacteria and fungi in

535 marine air-mass samples exhibited a positive correlation with air temperature,

536 suggesting elevated temperature in spring is conducive to the survival and reproduction

537 of bioaerosols. The present study demonstrated that air masses from different sources

538 would lead to different water-soluble ions, metal elements and microbial components

539 and concentrations in $PM_{2.5}$. This understanding will enhance our comprehension of the

540 environmental and climatic impacts on microbial aerosols within the marine boundary

541 layer. Our research offers a novel perspective on the variability of airborne microbial

542 communities and provides evidence suggesting that the atmospheric microbiome in

543 coastal cities is influenced by terrestrial and marine air masses. Further research is

544 required to conduct comprehensive studies on the sources, occurrence, and complex

545 amalgamation processes of coastal bioaerosols.

546 **CRediT authorship contribution statement**

547 All authors contributed to the manuscript and have given approval of the final version.

548 Min Wei designed the study. Qun He performed the data analysis and wrote the original

549 manuscript. Min Wei assisted with the sampling, Qun He, Zhaowen Wang and Rongbao

550 Duan conducted the experiments and performed the statistical analyses. Houfeng Liu,

551 Min Wei, and Pengju Xu contributed to the interpretation of results. Min Wei, Caihong

552   Xu and Jianmin Chen revised the manuscript.

Data availability

554   Hourly and daily average air quality and meteorological factor data were obtained
555   from   Shandong   Province   Ecological   Environment   Big   Data   Platform
556   (http://27.211.168.253:18102/portal/);   Meteolnfo   backward   trajectory   model
557   (MeteoInfo   3.7.4   -   Java,   http://www.meteothink.org   /downloads/index.html);
558   meteorological   data   were   obtained   from   GDAS1
559   (ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1/), and meteorological data as well as
560   bacterial and fungal concentrations are available from the authors on request
561   (minwei@sdnu.edu.cn). The original 16S rRNA and ITS gene sequences are available
562   in the Sequence Read Archive (SRA) under accession number PRJNA1096829.

Declaration of competing interest

564   The authors declare that they have no conflict of interests.

Acknowledgments

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