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

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Abstract:

Coastal environments provide an ideal setting for investigating the intermixing processes between terrestrial and marine aerosols. In this study, fine particulate matter (PM$_{2.5}$) samples were collected from a coastal location in Northern China and categorized into terrestrial, marine and mixed air masses. The chemical and biological constituents, including water-soluble ions (WSIIs), metallic elements, and bacterial and fungal aerosols, were investigated from January to March 2018. Terrestrial air masses constituted 59.94% of the total air masses at the sampling site throughout the period, with a significant increase during periods of severe haze pollution (up to 90%). These air masses exhibited higher concentrations of PM$_{2.5}$ (240 μg/m^3) and carried more water-soluble ions and metal elements. The terrestrial air mass also contained a greater number of animal parasites or symbionts, as well as human pathogens from anthropogenic emissions, such as *Staphylococcus*, *Deinococcus*, *Sphingomonas*, *Lactobacillus*, *Cladosporium* and *Malassezia*.

In contrast, saprophytic bacteria and fungi, including hydrocarbon-degrading bacteria and gut bacteria from *Comamonas*, *Streptococcus*, *Novosphingobium*, and *Aerococcus*, and *Aspergillus*, were the most prevalent species in the marine air mass. The mixed air mass revealed the intermixing processes of terrestrial and marine sources, resulting from microorganisms from anthropogenic and terrestrial emission, which including pathogenic microorganisms from hospitals and sewage treatment plants, and numerous soil bacteria. Correlation analysis indicated a stronger correlation between microorganisms and continental elements in terrestrial and mixed air mass samples, such as K^+, Mg^{2+}, and Ca^{2+} from soil dust. Marine air masses contain a significant quantity of saprophytic and intestinal microorganisms and exhibited a significant correlation with sea salt ions, specifically Na^+. In the mixed air mass sample, a fusion of marine and terrestrial microorganisms is characterized by alterations in the ratio of pathogenic to saprophytic microorganisms when compared to samples derived from either terrestrial or marine sources. This study on the constituents and amalgamation of bioaerosols over the coastal atmosphere encompassing distinct air masses is crucial for understanding the transport, intermixing processes, and health implications of terrestrial and marine air masses.

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

1. Introduction

Bioaerosols, encompassing bacteria, fungi, viruses, pollen, and cellular debris, etc., are vital aerosol particles in the atmosphere. Notably, bacterial aerosols which can be either free-floating or attached to airborne particles, typically measure between 0.3 and 10 μm (Shen and Yao, 2023; Zhao et al., 2022). Such characteristics allow for extended residence times and a heightened likelihood of long-distance transport from local to distant regions within the atmospheric circulation (Galbán et al., 2021; Smets et al., 2016). Fungal aerosols are prevalent in Earth's near-surface atmosphere, with their origins being diversely found in water, soil, and plants (Janine et al., 2009). Furthermore, the presence of pathogenic bacteria and fungi in the atmosphere may lead to significant penetration efficiency within the human respiratory system (Fakunle et al., 2021; Jiang et al., 2022).

Relevant studies have demonstrated that the bacterial and fungal concentration and community structure are significantly influenced by emission sources, atmospheric circulation, meteorological conditions, geographical and topographical factors (Liu et al., 2021; Núñez et al., 2021; Li et al., 2019). The geographical and topographical factors, such as terrestrial and marine environments exhibit significant differences in bioaerosol sources and pollution characteristics. Studies focusing on continental environments, particularly in large inland cities like Beijing (Zhang et al., 2019; Zhang et al., 2022), Xi'an (Wang et al., 2020; Yao et al., 2019), Jinan (Wei et al., 2020) , and Nanjing (Hu et al., 2020) during severe air pollution, have revealed that continental pollutant emissions significantly contribute to air pollution. Furthermore, transboundary transportation of terrestrial air masses on hazy days plays a crucial role in the progression of regional heavy pollution (Xie et al., 2020). The ocean serves as both a potential source and sink for airborne microorganisms (Archer et al., 2020;

Mayol et al., 2017), exhibiting the intricate interplay between continental communities and their transmission across oceanic regions (Cho and Hwang, 2011; Deleon-Rodriguez et al., 2013). Bioaerosols from the oceans maybe influenced by long-distance transport from continental sources, such as plants and human pathogens (Elbert et al., 2007; Sharoni et al., 2015). Studies have shown that the concentration and diversity of bacterial and fungal aerosols from marine are typically lower than those derived from continental sources (Cao et al., 2024; Xue et al., 2022; Shi et al., 2022). Xu et al. (2019) undertook a thorough investigation of bacterial abundance in Mt. Tai, China. Their findings indicate that variations in airmass from diverse sources could potentially influence the chemical composition of $PM_{2.5}$. This in turn prompts shifts in bacterial groups. Limited studies have examined the impacts of terrestrial and marine air masses on chemical constituents and microbial aerosols (Shi et al., 2022; Aswini and Hegde, 2021; Lang-Yona et al., 2022). Generally, the chemical aerosols can be affected by various sources of air masses, which may include local aerosols or remotely transported aerosols. However, it remains unclear whether different air trajectories contribute to the formation of bacterial communities within these particles. There is a dearth of research focused on determining the chemical and biological composition of coastal cities affected by these air masses.

Marine microbial aerosols can be released from ocean micro-surface or transported from land (Prospero et al., 2005) and settle thousands of kilometers away from their source of release (Mayol et al., 2014), exerting significant impacts on ecological and climate systems. Gong et al (2020a) conducted an examination of the microbial composition along Qingdao's coastlines, revealing a higher proportion of bacteria to total microorganisms in samples from continental sources compared to marine sources. Some marine bacteria persist in aerosols after land transportation, Xu et al

(2020b) studied the diversity of bacterial populations in PM$_{2.5}$ across urban and rural areas of Shanghai, finding that airborne microbial communities over coastal cities are more influenced by long-distance transport than those inland. Air samples taken at high altitude zones in coastal Europe and Japan demonstrated the continental transportation of marine microorganisms (Maki et al., 2014). The introduction of marine bacteria into tropospheric free space may modify the airborne microbial composition in continental regions (Cho and Hwang, 2011; Amato et al., 2007; Maki et al., 2014; Polymenakou et al., 2008; Cáliz et al., 2018). Consequently, it is imperative to ascertain the impact of terrestrial and marine air masses on the composition of atmospheric microorganisms, with a particular focus on the influence of the mixing process between terrestrial and marine air masses on bioaerosols. The costal aerosols provide the ideal conditions for understanding the mixing processes taking place between natural and anthropogenic air masses from terrestrial and marine.

Weihai, a coastal city situated at the confluence of eastern Asia continent and the Northwest Pacific Ocean, is prone to the impact of marine and terrestrial air masses. Typically, Weihai maintains low pollutant emissions with an annual average PM$_{2.5}$ concentration below 35 μg /m^3. However, in winter and spring, regional air pollution intensifies, leading to severe air quality issues. This is due to increased inter-regional transportation, resulting in daily average PM$_{2.5}$ concentration greater than 150 μg/m^3 (Wei et al., 2020). In this study, we conducted an integrated atmospheric observation experiment to examine the potential impact of terrestrial and marine air-mass on the constituents and amalgamation of bioaerosols over the coastal atmosphere. This approach allows for a comprehensive exploration of the effects of sea-land air mass exchange on the spatial and temporal distribution of aerosols, as well as potential intermixing processes in coastal regions.

2. Materials and methods

2.1 Sample collection

The sampling site was situated at the national air sampling station (37.53°N, 122.06°E), 41 m above sea level, and approximately 1-2 km from the coast, (Fig. S1). The sampling platform was about 15 m above ground level, and devoid of any significant obstructions. $PM_{2.5}$ samples were gathered between January to March, 2018, during the winter heating and spring dust seasons in northern China. Two parallel $PM_{2.5}$ samplers (TH-150C-III, Wuhan Tianhong Instrument Co., Ltd., China) with a particle size of 2.5 ± 0.2 μm were utilized equipped with Quartz membrane for the collection of $PM_{2.5}$, inorganic ion, metal elements, and microorganisms at a flow rate of 100 L min^{-1}. 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 (7:00~18:30 and 19:00~06:30 the following day). During the sampling process, the pre-weighed filter membrane was carefully positioned onto the filter mesh within a clean sampling folder using tweezers. The membrane's rough side should be oriented towards the direction of air intake, and it should be firmly pressed until there is no detectable air leakage. Prior to membrane replacement, the membrane tray should be wiped with alcohol cotton and dried. After sampling, it should be removed using tweezers. Blank samples were procured by putting the membrane into the sampler head without initiating the sampler. After sampling, the blank samples were extracted simultaneously with the collected sample membrane. All samples were then placed in a sterile filter cassette, encased in clean aluminum foil, and stored at -80°C until further analysis. During the whole sampling period from January 1 to March 31, a total of 102 $PM_{2.5}$ samples over 51 days were

143   obtained, out of which 24 samples were chosen for subsequent analysis based on

144   air mass categories. Meteorological parameters, such as air temperature, relative

145   humidity, wind direction, and wind speed, were monitored in situ utilizing a PC-4

146   automatic weather station (PC-4, JZYG, China). The hourly concentrations of $PM_{2.5}$,

147   PM_{10}, CO, SO_2, NO_2, and O_3 were systematically retrieved from the National Ambient

148   Air Quality Monitoring System (http://www.cnemc.cn/).

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

150   quantified after sampling. The membranes were meticulously weighed utilizing a

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

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

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

154   autosampler Thermo Fisher) was employed to determine the concentration of water-

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

156   extracted by sonication with ionized water for 45 min and then separated by anion

157   (IonPacAS23) or cation column exchange (IonPacCS12A). Then, these ions were

158   detected using a conductivity detector with an anion separation column of IonPacAS23,

159   flow rate: 1.0 mL/min, an anion suppressor of AERS500, and conductivity detector. The

160   injection volume was 25 μL, and the cation separation column was IonPacCS12A, flow

161   rate:1.0 mL/min, cation suppressor was CERS500 (Zhang et al., 2022). Metallic

162   elementals including Al, Fe, Ti, Mn, Co, Ni, Cu, Zn, Ga, Sr, Cd, Sn, Sb, Pb, V, Cr, and

163   As, were extracted using microwave digestion extraction (ETHOS ONE, Milestone),

164   with the concentrations determined by ICP-MS or ICP-OES (Thermo Fisher).

165   **2.2 Air mass clustering and classification**

166   The potential sources and transportation of air mass were examined using the

167 MeteoInfo backward trajectory model (MeteoInfo 3.7.4 – Java,

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

169 of Meteorological Sciences. Backward trajectories were simulated one-hour intervals

170 and estimated over a 24-hour period. The meteorological data were sourced from

171 GDAS1 (ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1/). Backward trajectories of air

172 masses at an altitude of 500 meters were categorized and clustered, with daily plots

173 illustrating these trajectories. In this study, air mass categories were defined that if more

174 than 90% of the masses originated from the ocean it was a typical sample affected by

175 marine air-mass; a typical terrestrial air-mass sample if more than 90% originated from

176 the continent; and mixed air-mass sample if the proportions of terrestrial and marine air

177 masses were similar or accounted for more than 40% of the total air masses in one day.

178 **2.3 DNA extraction and qPCR amplification**

179 Microbial genomic DNA were procured from filters utilizing the Fast-DNA^{TM} SPIN

180 kit for soil (MoBio Laboratories, Carlsbad, CA, USA). The concentration and quality

181 of the extracted DNA was measured via a Nanodrop spectrometer (Nanodrop 2000,

182 Thermo Scientific USA). The DNA concentration in the collected samples ranged from

183 10 to 20 ng/μL, satisfying the prerequisites for subsequent PCR amplification. The

184 quantitative polymerase chain reaction (qPCR) was employed to identify bacterial 16S

185 rRNA and fungal ITS gene copy numbers, as well as to estimate the count of bacteria

186 and fungi per cubic meter of air. The bacterial 16S V3-V4 variable region was selected

187 for PCR amplification using primer 338F (5'-ATCTACGGGGGGCAGCAG-3') and

188 806R (5'GGACTACHVGGGTWTCTAAT-3') (Masoud et al., 2011). The fungal ITS

189 region were amplified using the primers ITS1 (5′-CTTGGTCATTTAGAGGAAGTAA-

190 3′) and ITS2 (5′-GCTGCGTTCTTCATCGATGC-3′) (Liu et al., 2021).

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

for 5 minutes, succeeded by 30 seconds at 95°C, 30 seconds at 50°C, and then 35 cycles

at 72°C for 40 seconds, followed by a final extension at 72°C for seven minutes to

ensure comprehensive amplification. Fluorescent signals were gathered during this

extension phase. For each sample, qPCR was conducted in triplicate, with ultrapure

water serving as a negative control to identify potential PCR contamination. Standard

curves were constructed using *E. coli* harboring the 16S rRNA gene and *Streptomyces*

plasmids containing the ITS gene. Additionally, gradient dilutions of these plasmids

were performed, ranging from 10^2-10^7 copies/μL. The FTC-3000 real-time quantitative

PCR system was employed for standard curve construction and data processing.

2.4 16S rRNA and ITS gene sequencing and data processing

The V3-V4 region of the bacterial 16S rRNA and the fungal ITS1 gene were targeted

for PCR amplification utilizing barcode-specific primers 338F-806R and ITS1F-ITS2,

respectively. To ensure optimal amplification efficiency and precision, a high-fidelity

enzyme (Phusion® High Fidelity PCR Master Mix from NewEngland Biolabs) in

conjunction with GC buffer was employed during PCR amplification.

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

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

seconds; and 72°C for an additional 30 seconds, with a final extension at 72°C for five

minutes. Following amplification, samples were purified using the Agencourt Ampure

XP kit (from Beckman Coulter, Brea, CA, USA). Subsequently, the purified samples

were combined to achieve equimolar concentrations and analyzed on the Illumina

MiSeq PE300 platform (Illumina, Inc. in San Diego, CA).

Following sequencing, the barcode sequences were extracted and subsequently

stored in fastq format utilizing the QIIME toolkit (Caporaso et al., 2010). Sequences shorter than 200 base pairs, with mass of less than 25, and containing ambiguous bases underwent quality control screening using Trimmomatic (Bolger et al., 2014) and Mothur (Schloss et al., 2009). The assembled sequences were then de-duplicated and trimmed to equal length. De-chimerized sequences were eliminated (Edgar, 2013), and sequences were clustered into operational taxonomic units (OTUs) using UPARSE at a 97% identity threshold. Subsequently, individual OTUs were removed. Taxonomic assignments were determined using a Basic Local Comparison Search Tool (BLAST) search against the bacterial SILVA taxonomy release 138.2 dataset and fungal UNITE ITS reference v8.0 dataset. The original raw sequences were deposited in the Sequence Read Archive under the accession number PRJNA1096829.

2.5 Statistical analysis

Bacterial community functional was conducted using FAPROTAX, a manually constructed database that maps prokaryotic taxa to metabolic or other ecologically functions, such as sulfur, nitrogen, hydrogen, and carbon cycling (Chen et al., 2022). FUNGuild (Fungi Functional Guild) was used to predict the fungal ecological function. This tool could classify and analyze fungal communities by the microecological guild based on current published literature or data from authoritative websites to classify fungi functionally (Nguyen et al., 2016). Three primary groups are obtained based on the nutritional mode: Pathorotroph, Symbiotroph, and Saprotroph. Samples affected by different airmasses were examined for intergroup species variability, based on community abundance data. This was achieved using rigorous statistical methods to identify species demonstrating differences in abundance within the microbial communities of different groups, and hypothesis testing to evaluate the significance of

239 these observed differences. Statistical analysis, including Analysis of variance

240 (ANOVA) and Kruskal Wallis tests, were employed to discern bacteria and fungi with

241 varying abundances between samples and groups. A *p* value less than 0.05 or 0.01 was

242 considered significant. ANOVA was used to analyze variation in a response variable

243 measured under conditions defined by discrete factors (Martin G. Larson, 2008). The

244 Kruskal-Wallis test determines whether there is a statistically significant difference

245 between the medians of three or more independent groups (Kassambara, 2019). The

246 Mantel analysis was utilized to reveal the correlation between microbial community

247 and various environmental factors. The Pearson's r coefficient at $p<0.05$ and $p<0.01$

248 indicates the significant correlation.

249 **3. Results**

250 **3.1 Air masses categorization and typical pollution processes**

251   The Meteolnfo backward trajectory model was employed to simulate the trajectories

252 of air masses at an altitude of 500 m in Weihai over 24-hour period,

253 identifying and classifying potential sources of air mass transport (Fig. S1). The

254 terrestrial air mass accounted for 59.94% of the total air masses at the sampling

255 site throughout the period, exhibiting an average $PM_{2.5}$ concentration of 36.15±26.52

256 $\mu g/m^3$. Severe air pollution episodes occurred on January 20, with $PM_{2.5}$ was the

257 primary pollutant and the concentration reaching 240 $\mu g/m^3$. During reginal haze

258 pollution, the terrestrial air masses primarily influenced Weihai were typically

259 originating from the Beijing-Tianjin-Hebei region and the surrounding areas. The

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

261 population, industrial and agricultural activities. The impact of the spring dust is mainly

262 reflected in the primary pollutant PM_{10} on March 30. A significant increase in PM_{10}

263　concentration was observed on March 30, with an hourly maximum value of 197 μg/m^3.

264　A relatively low PM$_{2.5}$/PM$_{10}$ ratio of 0.28 indicated pronounced dust pollution.

265　　The typical marine air masses mainly from the eastern and southern areas from the

266　Yellow Sea and the western Pacific edge. Marine air masses from northern areas partly

267　come from the Bohai Sea, crossing inland areas and possibly making contact with the

268　land prior to reaching the study area (Fig. S1). A total of 14% of the sampling days were

269　influenced by marine air masses, resulting in an average PM$_{2.5}$ concentration of

270　23.99±11.00 μg/m^3. Mixed air masses, characterized by simultaneous influence from

271　the northwestern winds of Inner Mongolia and the offshore air masses of the Bohai Sea

272　or Yellow Sea, accounted for 27% of all affected sampling days. These mixed air masses

273　yielded an average PM$_{2.5}$ concentration of 45.11±12.69 μg/m^3. In comparison, under

274　the influence of mixed air masses, pollutant concentrations were notably high. During

275　the spring dust season, there is a notable increase in the proportion of mixed air mass.

276　This elevated concentration of particulate matter correlates with the transmission of

277　sand dust (Xie et al., 2020).

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

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

280　events were predominantly characterized by terrestrial and mixed air masses. Cold

281　northwestern airmass form the continent, and marine air masses were the primary

282　contributors during pollution mitigation. For pollution episode I occurred from January

283　19 to January 22, the western terrestrial air mass was predominantly responsible for

284　initiating the pollution, evidenced by a PM$_{2.5}$ concentration of 51.35 μg/m^3. As the

285　pollution progressed, it transitioned into a mixed air mass with a PM$_{2.5}$ concentration

286　peaking at 240 μg /m^3. This change was accompanied by significant increases in WSIIs

287　and elemental concentrations. Pollution elimination was initiated with high winds,

snowfall influenced by the cold northwestern airmass form the continent and marine air masses from the northeast sea. An average $PM_{2.5}$ concentration reduction to 7.92 μg /m^3, with lower WSIIs and elemental concentrations.
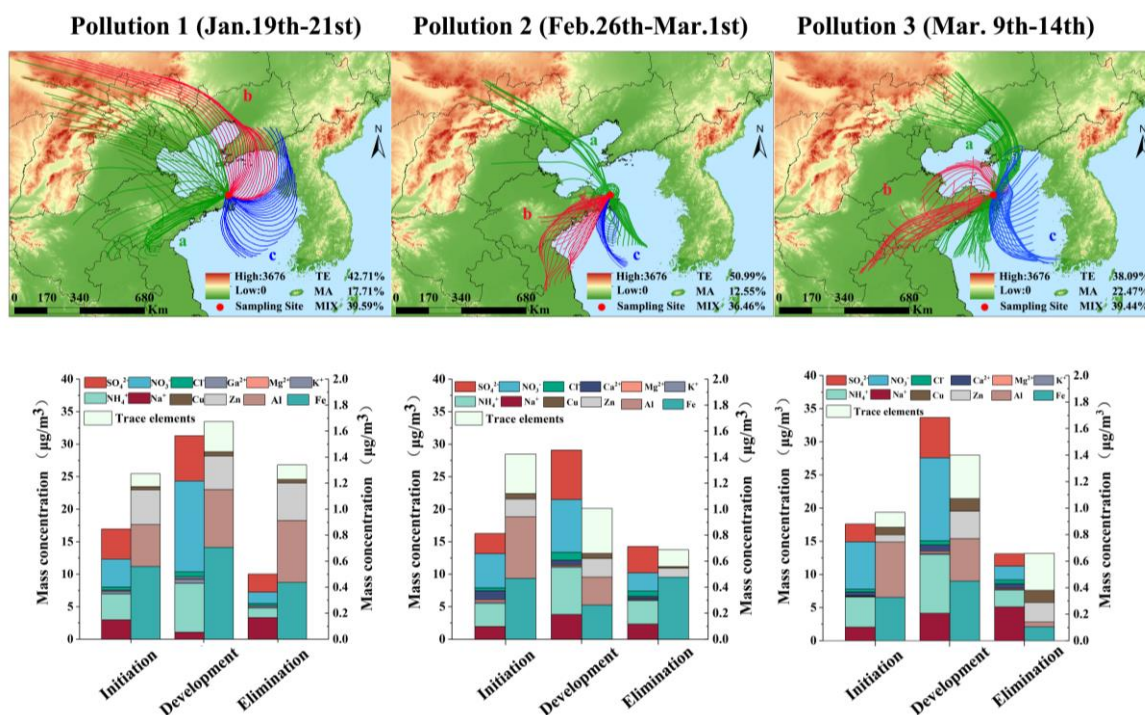
Fig. 1 Transformation of terrestrial and marine air masses, and chemical composition in $PM_{2.5}$ of three severe air pollution episodes. a, pollution initiation; b, pollution development; c, pollution elimination. TE, terrestrial air mass; MA, marine air mass; MIX, mix air mass.

3.2 Water-soluble ions and metal elements over coastal atmosphere

During the sampling period, the daily concentration of water-soluble ions in $PM_{2.5}$ was 22.92±12.19 μg/m^3, ranging from 4.58 to 78.14 μg/m^3. In marine air-mass samples, a 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. Notably, NO_3^- had the highest concentration (26.94%, 6.4%~52.6%), succeeded by SO_4^{2-} (21.94%, 9.4%~33.4%) and NH_4^+ (20.26%, 5.8%~35.6%). The concentration of SO_4^{2-}, NO_3^- and NH_4^+, constituted the majority of

total ion in other studies conducted in Beijing, Shanghai, and Guangzhou (Pathak et al., 2008; Hu et al., 2014; Zhou et al., 2012). These inorganic secondary ions 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).

A high concentration of Na^+ was observed, with a range that from 3.15 ± 1.69 μg/m^3, which accounted for 14.47% of the total water-soluble ion. The concentration of Na^+ and Mg^{2+} exhibited analogous trends across terrestrial, marine, and mixed air mass samples. The ratio of Mg^{2+}/Na^+, typical components of sea salt, was determined to be 0.11. This value closely corresponded to the 0.12 found in seawater, indicating a potential origin from marine environments (Sun et al., 2022). The concentration of K^+ 0.24 ± 0.20 μg/m^3 and 0.26 ± 0.10 μg/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 ± 0.05 μg/m^3), which suggested an important contribution from anthropogenic emissions. The concentration of Cl^-, and Ca^{2+} was high in terrestrial and mixed air mass samples. Generally, these two ions mostly come from anthropogenic or terrestrial environments, such as Cl^- mainly comes from sea salt, coal and biomass combustion, and Ca^{2+} is a crustal element associated with soil dust and sandstorms (Liang et al., 2021). Overall, from the composition and concentration of water-soluble ions in PM$_{2.5}$, the coastal city was more affected by sea salt, coal combination in winter heating season and dust events in early spring.

The elements concentration in different air-mass samples are depicted in Fig. S3. The top ten elements were found to be Fe, Al, Zn, Cu, Sn, Pb, Mn, Ti, Ni, and V. These metal elements were categorized into macro and trace elements. The macro metals, specifically Iron (Fe), Aluminum (Al), and Zinc (Zn), constituted a significant

329 proportion of the total heavy metal elements, accounting for 34%, 25%, and 23%

330 respectively. In general, the concentration of both macro and trace metal elements in

331 marine air masses was found to be lower than that in terrestrial and mixed air masses.

332 The V/Ni ratio in marine air-mass samples was found to be 0.78, a value significantly

333 higher than that of both terrestrial and mixed air masses. The V/Ni ratio is employed as

334 a measure of the influence of ship emissions. A ratio exceeding 0.7 typically indicates

335 a significant impact from these emission sources, and is commonly used as an indicator

336 in coastal cities (Zhang et al., 2014).

3.3 Microbial community over coastal atmosphere

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

339 (P=0.06) and $3.39\pm1.10\times10^4$ cells/m^3 (P=0.04), respectively. This value was

340 comparable to the mean abundance of bacterial and fungal spores in the atmospheric

341 boundary layer over land, which is approximately 1.9×10^4 cells/m^3 and 2.4×10^4

342 cells/m^3, respectively (Spracklen and Heald, 2014; Mayol et al., 2014). A high

343 concentration was observed in terrestrial air-mass samples, with the average value of

344 $4.72\pm3.93\times10^5$ cells/m^3 (P=0.12) and $6.37\pm1.70\times10^4$ cells/m^3 (P=0.007) for bacteria

345 and fungi, respectively. Microbial concentration in marine air-mass samples was

346 significantly lower, with the average concentrations of bacteria and fungi being

347 $4.91\pm1.82\times10^4$ cells/m^3 (P=0.04) and $6.15\pm3.09\times10^3$ cells/m^3 (P=0.07).

348 Airborne microbial community structure exhibited significant variation influenced

349 by marine and terrestrial air masses (Fig. 2). Predominantly, *Proteobacteria* (40.06%),

350 *Firmicutes* (36.30%), *Actinobacteria* (8.97%), *Bacteroidetes* (8.29%), and

351 *Deinococcus-Thermus* (4.59%) were identified as the most abundant bacteria. Notably,

352 *Actinobacteria* and *Deinococcus-Thermus* were found in high concentration in samples

from terrestrial air masses. In particular, *Deinococcus-Thermus* demonstrated a relative abundance of 7.9% in terrestrial air mass samples, significantly surpassing that of marine (1.2%) and mixed air mass samples (1.5%). *Cyanobacteria* exhibit a higher concentration in both terrestrial and mixed air-mass samples. *Cyanobacteria* are prevalent in freshwater, soil, surface crusts in deserts (Cordeiro et al., 2020; Temraleeva et al., 2016; Curren and Leong, 2020). In comparison to terrestrial environments, the prevalence of *Cyanobacteria* is notably diminished in colder marine environments (Koh et al., 2012). Atmospheric *Cyanobacteria* in the coastal city may originate from the terrestrial environment, subsequently transported via terrestrial and mixed air masses. *Proteobacteria* was the most prevalent taxon in marine air-mass samples. The predominant metabolic activity in deep-sea sediments is attributed to this group, with major taxa being found in marine sediments (Huang et al., 2021). The predominant bacterial genera included *Staphylococcus* (21.02%), *Enhydrobacter* (6.43%), *Lactobacillus* (6.03%), and *Deinococcus* (4.56%). These bacteria including a series of opportunistic pathogens and were found abundant in terrestrial and mixed air masses samples. Relative abundance of bacteria such as *Comamonas*, *Streptococcus*, *Ruminococcus*, and *Enterobacteriaceae* were higher in marine air-mass samples. These predominantly consisted of saprophytic bacteria and intestinal microorganisms.

The dominant fungal phyla were *Ascomycota* (77.29%) and *Basidiomycota* (21.58%), which were similar to the previously studies (Du et al., 2018; Liu et al., 2019; Zeng et al., 2019). Fungal community influenced by terrestrial and mixed air masses were quite similar, with relatively higher abundances of opportunistic pathogens such as *Malassezia*, *Alternaria*, *Cladosporium*. In contrast the saprophytic *Aspergillus*, *Davidiellaceae* and *Coprinellus* were abundant in marine air-mass samples.
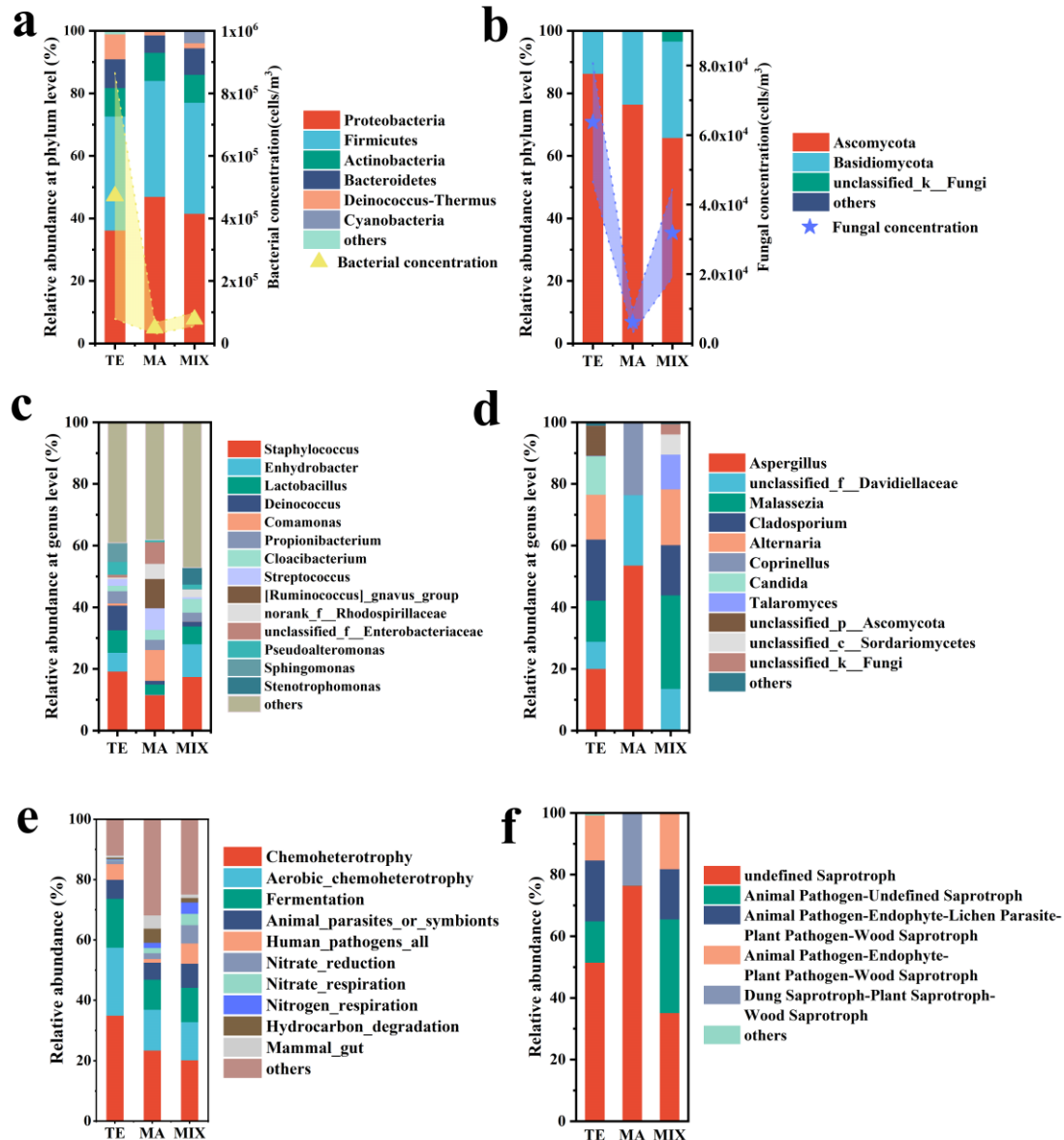
Fig. 2. Bacterial and fungal community composition and function influenced by different air masses. The community concentration, main phylum (a), (b), genus (c), (d), and community function (e), (f) are indicated.

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. Marine air-mass samples were enriched with mammal gut bacteria,

as well as hydrocarbon and automatic compound degradation bacteria. The fungal community function in terrestrial and mixed air-mass samples were similar, with undefined Saprotroph, Animal Pathogen-Undefined Saprotroph, Animal Pathogen-Endophyte-Lichen Parasite, Animal Pathogen-Endophyte-Plant Pathogen were the main functions, which totaled 99.27%, 99.98%, 99.27% in the terrestrial, marine, and mixed air-mass samples. In particular, the prevalence of Saprotroph fungi was observed higher in samples from marine air masses, such as those containing *Aspergillus*. Notably, the fungi associated with terrestrial air masses predominantly carried animal pathogens and exhibited greater pathogenicity, including species such as *Malassezia* and *Alternaria*.

4. Discussion

4.1 Community disparities influenced by terrestrial and marine air masses

Overall, bacterial and fungal community concentration influenced by terrestrial and mixed air masses were higher than those in marine air mass samples. Bioaerosols originating from anthropogenic environments, such as sewage treatment plants and hospitals, may harbor more resistant bacteria. Microorganisms from arid areas like deserts possess a higher number of species adapted to harsh conditions, including resistance to temperature fluctuations, dryness, and UV radiation. These include bacteria such as *Bacillus*, *Streptococcus*, *and Deinococcus* ((Maki et al., 2010; Park et al., 2018; Qi et al., 2021). These microorganisms can adhere to particulate matter during long-distance transport of airmasses after being released from their terrestrial habitats.
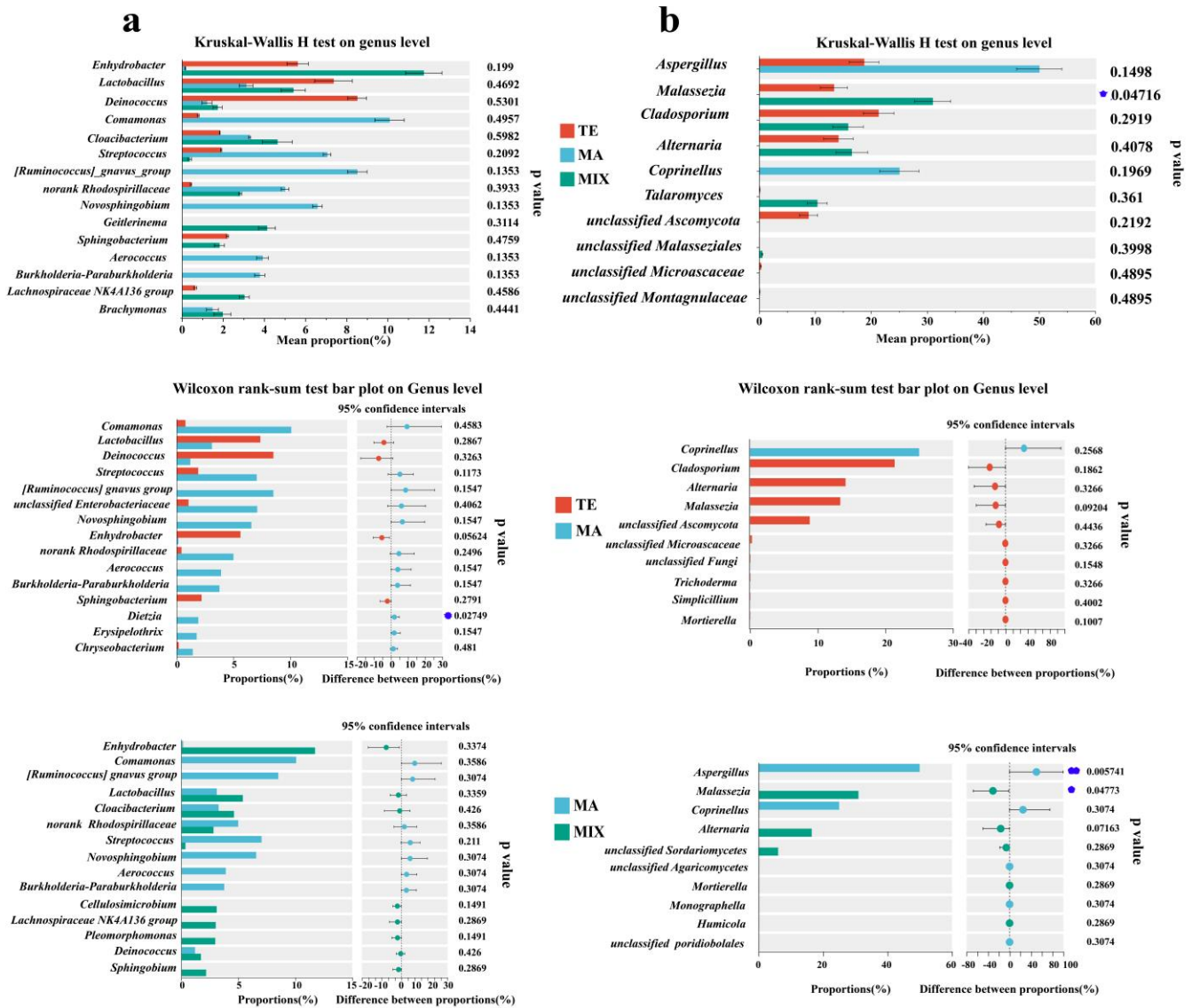
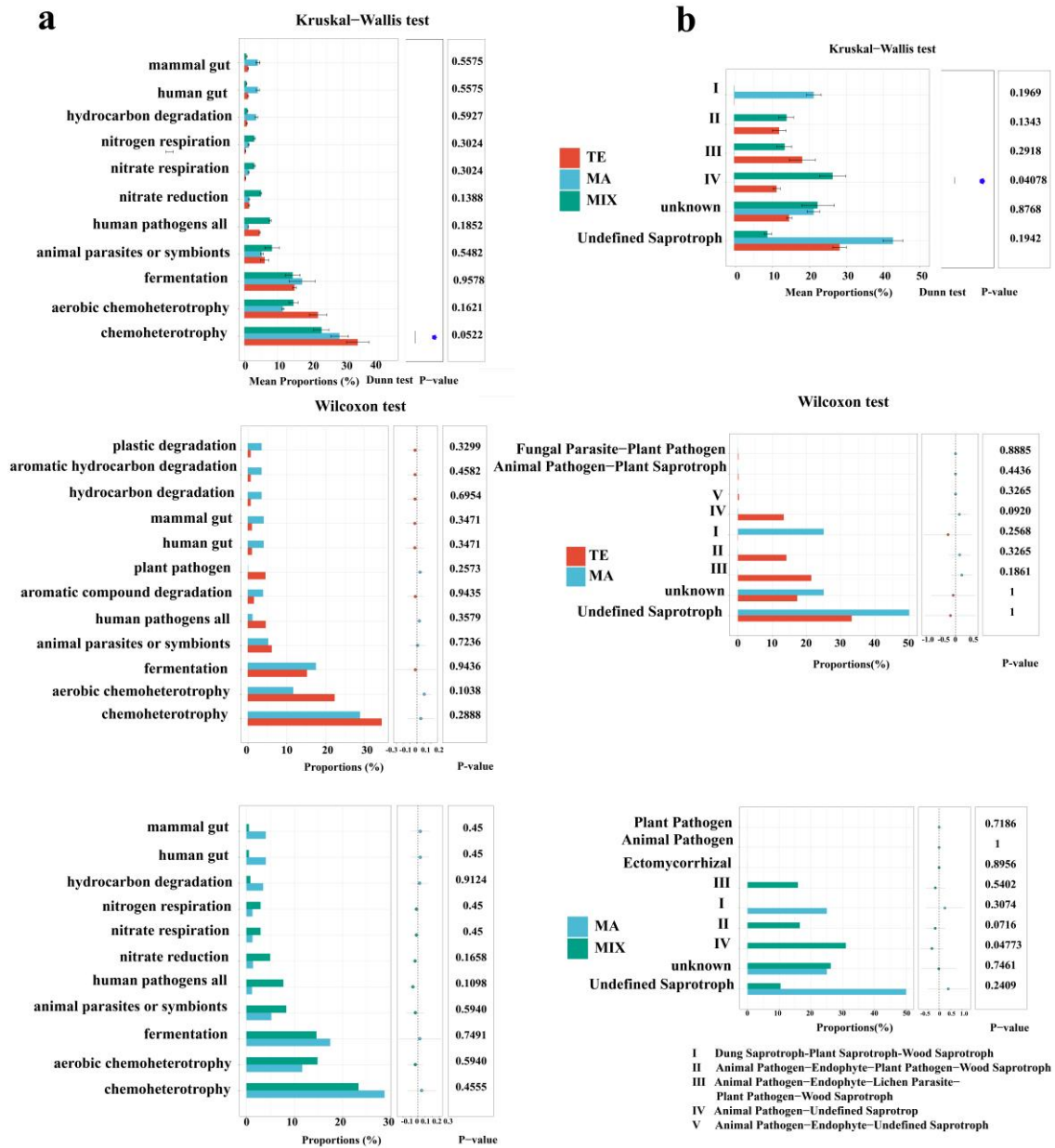**Fig. 3 Bacterial (a) and fungal (b) community disparities influenced by terrestrial (n=10), marine (n=6), and mixed mass (n=8).**

Community disparities influenced by terrestrial, marine, and mixed air masses was conducted in Table S2, S3, Fig. 3 and Fig. 4. The Principal Coordinates Analysis (PCoA) revealed distinct clusters corresponding to terrestrial, marine, and mixed air mass (Fig. S4). In terms of bacterial community variations, numerous dominant microorganisms exhibited obvious differences in abundance between marine and terrestrial air mass samples. However, these differences were not statistically significant (p>0.05). For instance, the bacterial enrichments in terrestrial and mixed air-masses included *Staphylococcus, Deinococcus*, *Lactobacillus*, and *Sphingomonas*. *Staphylococcus is* a

pathogenic bacterium widely found in human skin and intestines that can cause various infections (Cheung et al., 2021). As a radiation-resistant bacterium, *Deinococcus-Thermus* is capable of withstanding harsh environmental conditions (Callegan et al., 2008; Rainey et al., 2007). *Lactobacillus*, a genus of Gram-positive bacteria, has also been identified as abundant in atmospheric dust (Federici et al., 2018; Xu et al., 2017). *Sphingomonas*, typically found in water and soil, which is widely known as the metabolic mechanism able to endure poor nutrients and degradation of refractory compounds, such as polycyclic aromatic hydrocarbons (PAHs) (Hu et al., 2007; Sun et al., 2018). The majority of these bacteria possess the ability to produce spores, which are capable of withstanding harsh conditions such as low temperatures, aridity, and radiation during long-distance transmission processes, thereby ensuring their survival throughout this process (Griffin et al., 2003). *Comamonas* was identified as the dominant bacterium in the coastal atmosphere of Weihai, particularly prevalent in marine air-mass samples. This bacterium is commonly associated with environmental bioremediation and is predominant in oligotrophic environments (Yan et al., 2012; Zhang et al., 2024). *Streptococcus* are mostly found in the oral and gastrointestinal tracts and also play an important role in human infections (Brouwer et al., 2023). *Ruminococci*, and *Enterobacteriaceae* are typical gut microorganisms present in humans and animals. In the coastal atmosphere, the identified sequences of these gut bacteria might be associated with intestinal microorganisms in marine fish and mammal animals.

For fungal community, *Aspergillus* demonstrated a significant differentiation between marine and mixed airmasses (P=0.005). The highest proportion was noted in samples from marine air masses, at 53.7%. In contrast, the values were 20.1% and 0.3% in terrestrial and mixed air masses respectively. Previous studies have confirmed that

Aspergillus is widely distributed in nature and unpolluted environments (Kendrick, 1995). This fungus is predominantly found in offshore regions, such as Qingdao, China (Li et al., 2011). Furthermore, the saprophytic Aspergillus was also prevalent in clean samples during periods of haze pollution and was frequently detected on non-haze days (Yan et al., 2016). *Malassezia* was higher in terrestrial and mixed air-mass samples (P=0.047), which has been found to be widespread in a variety of animals. As a parasitic fungus, *Malassezia* causes the majority of skin diseases, such as dandruff and seborrheic dermatitis caused by *Malassezia sphericalis* (Deangelis et al., 2007). Furthermore, *Cladosporium* exhibits higher abundance in terrestrial and mixed air-mass samples compared to marine air masses, although this difference lacks statistical significance (P>0.05). This species is ubiquitous worldwide, commonly found in a wide variety of plants, known to be a common endophyte as well as a foliar fungus (El-Morsy, 2000; Islam and Hasin, 2000). It is frequently isolated from soils, foodstuffs and organic matters (Bensch et al., 2012). Moreover, *Cladosporium* have been commonly observed in terrestrial atmospheric environments, demonstrating the potential origins from continental environments (Frączek et al., 2017; Han et al., 2019).

The significant variance in the ecological function of the microbial community was predominantly evident in the IV group of fungi, which are classified as Animal Pathogen-Undefined Saprotrophs (*P=0.04*). These pathogenic fungi were more prevalent in terrestrial and mixed air-mass samples than in marine air-mass samples. Marine air-mass samples were enriched with mammal gut bacteria, hydrocarbon and automatic compound degradation bacteria, and undefined Saprotroph fungi.

a

Kruskal−Wallis test

	Mean Proportions (%)	Dunn test P-value
mammal gut		0.5575
human gut		0.5575
hydrocarbon degradation		0.5927
nitrogen respiration		0.3024
nitrate respiration		0.3024
nitrate reduction		0.1388
human pathogens all		0.1852
animal parasites or symbionts		0.5482
fermentation		0.9578
aerobic chemoheterotrophy		0.1621
chemoheterotrophy		0.0522

b

Kruskal−Wallis test

TE MA MIX

	Mean Proportions(%)	Dunn test P-value
I		0.1969
II		0.1343
III		0.2918
IV		0.04078
unknown		0.8768
Undefined Saprotroph		0.1942

Wilcoxon test

	Proportions (%)	P-value
plastic degradation		0.3299
aromatic hydrocarbon degradation		0.4582
hydrocarbon degradation		0.6954
mammal gut		0.3471
human gut		0.3471
plant pathogen		0.2573
aromatic compound degradation		0.9435
human pathogens all		0.3579
animal parasites or symbionts		0.7236
fermentation		0.9436
aerobic chemoheterotrophy		0.1038
chemoheterotrophy		0.2888

Wilcoxon test

TE MA

	Proportions(%)	P-value
Fungal Parasite−Plant Pathogen		0.8885
Animal Pathogen−Plant Saprotroph		0.4436
V		0.3265
IV		0.0920
I		0.2568
II		0.3265
III		0.1861
unknown		1
Undefined Saprotroph		1

	Proportions (%)	P-value
mammal gut		0.45
human gut		0.45
hydrocarbon degradation		0.9124
nitrogen respiration		0.45
nitrate respiration		0.45
nitrate reduction		0.1658
human pathogens all		0.1098
animal parasites or symbionts		0.5940
fermentation		0.7491
aerobic chemoheterotrophy		0.5940
chemoheterotrophy		0.4555

MA MIX

	Proportions(%)	P-value
Plant Pathogen		0.7186
Animal Pathogen		1
Ectomycorrhizal		0.8956
III		0.5402
I		0.3074
II		0.0716
IV		0.04773
unknown		0.7461
Undefined Saprotroph		0.2409

I   Dung Saprotroph-Plant Saprotroph-Wood Saprotroph
II   Animal Pathogen−Endophyte−Plant Pathogen−Wood Saprotroph
III Animal Pathogen−Endophyte−Lichen Parasite−
     Plant Pathogen−Wood Saprotroph
IV   Animal Pathogen−Undefined Saprotrop
V   Animal Pathogen−Endophyte−Undefined Saprotroph

468

469 **Fig. 4. Bacterial (a) and fungal (b) community function disparities influenced by**

470 **terrestrial (n=10), marine (n=6), and mixed mass (n=8).**

4.2 Implications of environmental factors on coastal airborne microbes

To elucidate the contributions of marine and terrestrial air masses to microbial aerosols in the coastal city, a Mantel correlation analysis was conducted between microbial aerosols and various environmental factors (Fig.5, Fig.S4). The bacterial community exhibited a significant positive correlation with SO_4^{2-} due to the influence of terrestrial and mixed air masses ($P<0.05$). Both bacterial and fungal communities demonstrated positive correlations with continental ions such as K^+, Mg^{2+}, and Ca^{2+} (Shi et al., 2022). Additionally, a positive correlation was observed between bacterial and fungal concentrations and NO_2 ($P<0.05$), as well as a significant positive correlation with PM_{10} ($P<0.01$).

Air masses transported over long distances from the continent appear to harbor diverse and abundance microbial populations (Kakikawa et al., 2009; Deleon-Rodriguez et al., 2013). For instance, the long-distance transportation of dust particles from the northwestern winds in the Inner Mongolia region may have changed the community structure and abundance (Squizzato and Masiol, 2015; Castañer et al., 2017). Dust-borne bacteria (*Staphylococcus*, *Delftia*, *Pseudoalteromonas* and *Deinococcus*) were injected into the atmosphere during dust events, and most of them accompanied the dust transportation to the downwind of Asian Dust including the coastal city of Weihai. Influenced by mixed air masses, bacterial community was significantly positively correlated with K^+ (*P<0.01*) and PM_{10} (*P<0.05*).

Similarly, microbial communities showed high positively correlated with ions from continental sources, such as K^+, Mg^{2+}, and Ca^{2+} in terrestrial and mixed air mass samples. K^+ is the signature ion of biomass combustion (Mason et al., 2016; Yu et al., 2018). Mg^{2+}, and Ca^{2+} are mostly derived from crustal elements (Zhang et al., 2012). The dominant bacteria within the microbial community exhibited a significant positive

correlation with these three ions simultaneously, suggesting a similar sources. They

generally originate from terrestrial sources, including anthropogenic biomass

combustion and soil dust.
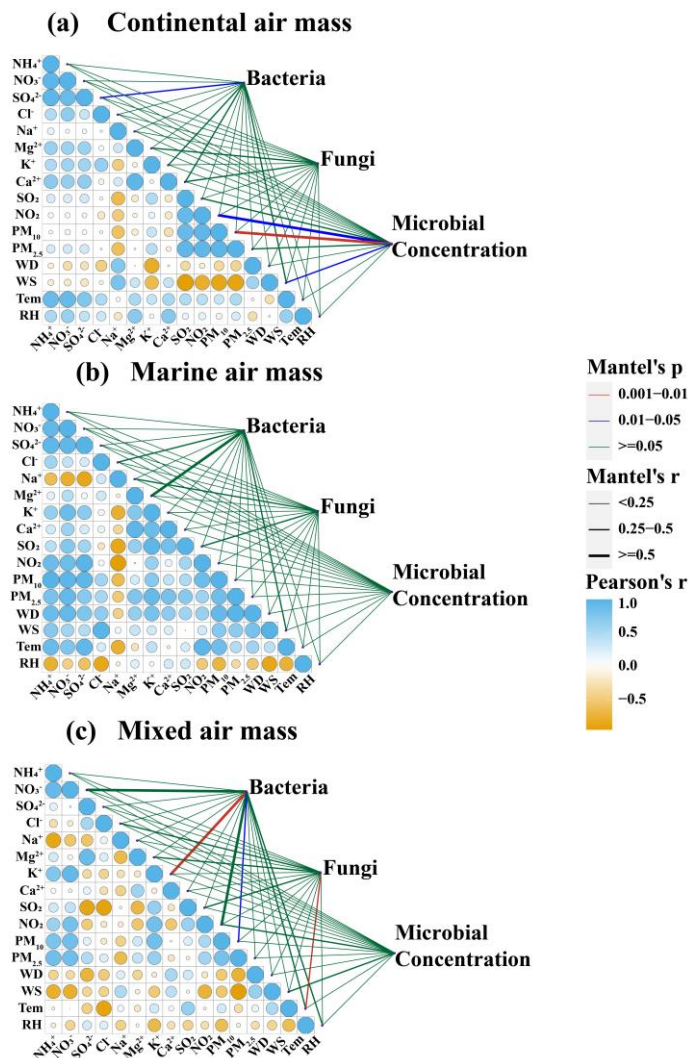
Fig.5 Mantel analysis reveal the correlation between microbial community and various environmental factors under different air masses, terrestrial air masses (a) marine air masses (b) and mixed air masses (c). The Pearson's r coefficient indicates the significant correlation at p<0.05 and p<0.01.

The concentrations of particulate matter and water-soluble ions in samples

influenced by the marine air mass are notably low, suggesting that most chemical

elements did not significantly impact the composition and abundance of the microbial

community. However, a significant positive correlation was observed between Na^+ and

509　certain dominant species such as *Talaromyces*, *Monographella*, and *Phoma* ($P < 0.05$).

510　Similarly, Mg^{2+} was found to be significantly positively correlated with *Malassezia* in

511　mixed air masses ($P<0.01$) (Fig. S5). Except for anthropogenic sources, such as

512　industrial emissions, the origins of Mg^{2+} and Na^+ in coastal regions should also consider

513　the impact of sea salt (Sun et al., 2022). Samples collected under the influence of marine

514　air masses had a moderate correlation between Mg^{2+} and Na^+ ($r^2 = 0.67$), indicating that

515　they have a similar origin of sea salt. Moreover, the average Mg^{2+}/Na^+ ratio was 0.11,

516　close to the value of 0.12 in seawater (Seinfeld and Pandis, 1997).

517　　Influenced by mixed air mass, temperature have a greater impact on fungal

518　community, which was positively correlated with *Malasseziales* and *Davidiellaceae*. In

519　marine airmass samples, a positive correlation between air temperature and certain

520　microorganisms (*Aerococcus*, *Cloacibacterium*, *Sphingobium*, *Enhydrobacterium*,

521　*Davidiellaceae*, *Malasseziales*) also indicated that the increase in air temperature in

522　spring favors the survival of airborne microbes (Jones and Harrison, 2004).

523　4.3 Intermixing of bioaerosols from terrestrial and marine air-mass

524　　Coastal atmospheres exhibit a complex amalgamation of terrestrial and marine

525　aerosols, with the characteristics significantly influenced by the origins of the air

526　masses. Consequently, these coastal aerosols offer ideal conditions for examining the

527　mixing processes between natural and anthropogenic air masses from terrestrial and

528　marine sources, respectively. This work shows that enormous levels of haze aerosols

529　from the terrestrial and mixed air mass can be rapidly transported into the coastal city

530　during regional haze pollution. During the long-distance transportation of air masses, a

531　comprehensive mixture of soil-derived, biogenic, and anthropogenic microorganisms

532　from terrestrial air masses, and aquatic, saprophytic, gut microorganisms from marine

environments are fully mixed (Fig. 6). 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 mixed air mass. The primary influence on terrestrial air mass was anthropogenic emissions, with coal combustion for winter heating and biomass burning being the predominant pollution sources. Moreover, dust events in spring carried higher concentrations of particulate matter. These air pollutants can act as initial source of bioaerosols such as bacteria and fungi, thereby providing a site for attachment reproduction (Jiang et al., 2022). Additionally, water-soluble ions in PM, primarily secondary ions, sulfate, nitrate, and ammonium ions, can supply essential nutrients for microbial growth (Fan et al., 2019). This explanation elucidates the increased concentration of particulate matter and microorganisms in terrestrial air mass during heavy pollution. Simultaneously, it is pertinent to highlight that during pollution incidents, the terrestrial air mass intensifies the pollution process. This intensification results in a significant increase in the proportion of pathogenic microorganisms.
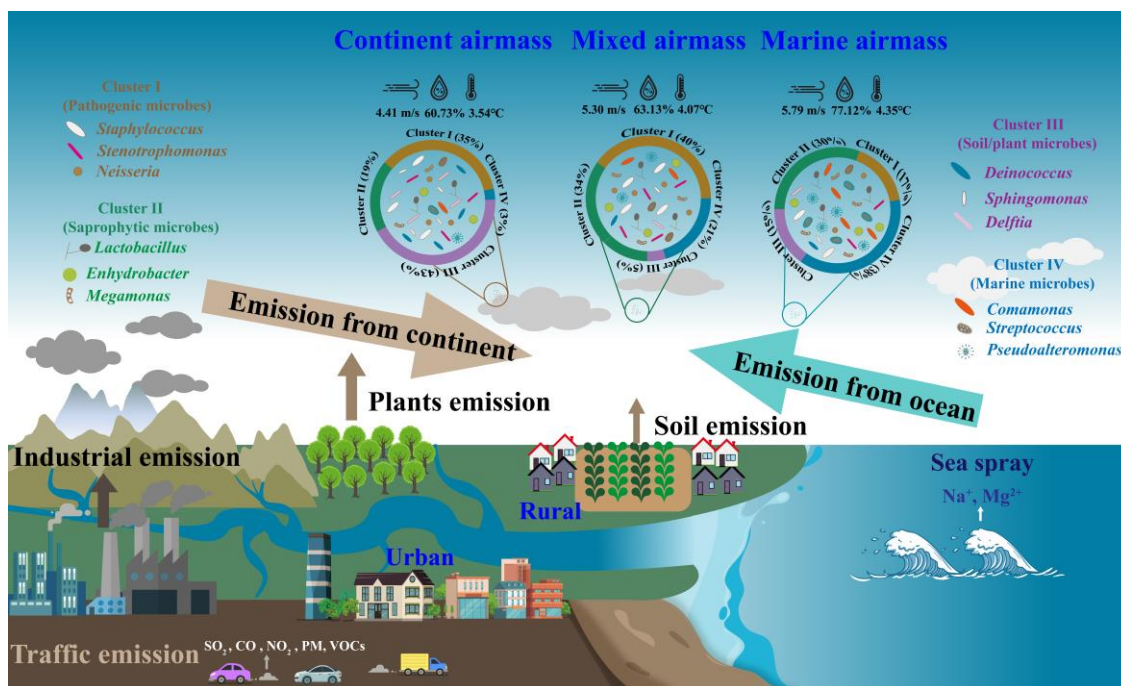
Fig. 6 Implications of intermixing of bioaerosols from terrestrial and marine air-mass on the composition of airborne microbial communities.

In the coastal city, marine air masses predominantly originate from the expansive western Pacific Ocean, located to the eastern and southern areas. These air masses are typically characterized by their cleanliness and contain a significant quantity of saprophytic and intestinal microorganisms that infiltrate the atmosphere via ocean droplets (Mayol et al., 2014). The presence of microorganisms in marine air masses exhibited a significant correlation with sea salt ions, specifically Na^+ and Mg^{2+}. These ions, when introduced into the atmosphere, form aerosolized particulate matter. Marine bacteria can be introduced into tropospheric free space may modify the airborne microbial composition in continental regions (Cho and Hwang, 2011; Amato et al., 2007; Maki et al., 2014; Polymenakou et al., 2008; Cáliz et al., 2018), exerting significant impacts on bioaerosols over coastal atmosphere.

Notably, marine air masses from the northerly areas, traverse China's inland Bohai Sea and subsequently pass through urban and rural regions with high human activity levels. Consequently, these areas introduce anthropogenic emission source

microorganisms, including pathogenic microorganisms from hospitals and sewage treatment plants. In the present study, samples within this category are identified as those influenced by a mixed air mass. This also signifies the amalgamation of marine and terrestrial microorganisms. Consequently, within the mixed air mass samples, the prevalence of pathogenic and saprophytic microorganisms is comparable. This similarity underscores the complex impact of both terrestrial and marine sources. Populations in coastal cities may also be susceptible to exposure to these bioaerosols and pathogens, which are transported over long distances during regional air pollution.

5. Conclusion

This study evaluated the potential influence and the mixing effects 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. 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 *Staphylococcus*, *Malassezia* and *Alternaria.* In contrast, marine air mass carried more gut and 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 correlated with SO_4^{2-}, K^+, Mg^{2+}, and Ca^{2+} and PM_{10} influenced by terrestrial and mixed air masses from continental sources. In contrast, influenced by marine air masses, bacterial and fungal community were strongly correlated with sea salt ions, such as Na^+. The effects of

meteorological factors on bioaerosols vary significantly under different dominant air masses. The predominant bacteria and fungi in marine and mixed air-mass samples exhibited a positive correlation with air temperature, suggesting elevated temperature in spring is conducive to the survival and reproduction of bioaerosols. Our research offers a novel perspective on the variability of airborne microbial communities and provides evidence suggesting that the atmospheric microbiome in coastal cities is influenced by terrestrial and marine air masses. This understanding will enhance our comprehension of the environmental and climatic impacts on microbial aerosols within the marine boundary layer. Future research endeavors will concentrate on the local emission and long-distance transport, as well as the contributions of both terrestrial and marine air masses on bioaerosol components in coastal cities.

CRediT authorship contribution statement

All authors contributed to the manuscript and have given approval of the final version. Min Wei designed the study. Qun He performed the data analysis and wrote the original manuscript. Min Wei assisted with the sampling, Qun He, Zhaowen Wang and Rongbao Duan conducted the experiments and performed the statistical analyses. Houfeng Liu, Min Wei, and Pengju Xu contributed to the interpretation of results. Min Wei, Caihong Xu and Jianmin Chen revised the manuscript.

Data availability

Hourly and daily average air quality and meteorological factor data were obtained from Shandong Province Ecological Environment Big Data Platform (http://27.211.168.253:18102/portal/); MeteoInfo backward trajectory model (MeteoInfo 3.7.4 - Java, http://www.meteothink.org /downloads/index.html);

meteorological data were obtained from GDAS1 (ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1/), and meteorological data as well as bacterial and fungal concentrations are available from the authors on request (minwei@sdnu.edu.cn). The original 16S rRNA and ITS gene sequences are available in the Sequence Read Archive (SRA) under accession number PRJNA1096829.

Declaration of competing interest

The authors declare that they have no conflict of interests.

Acknowledgments

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