

Role of aerosol liquid water content on the production of dicarboxylic acids in the dust-laden air masses over the Arabian Sea: Implications for heterogeneous chemistry

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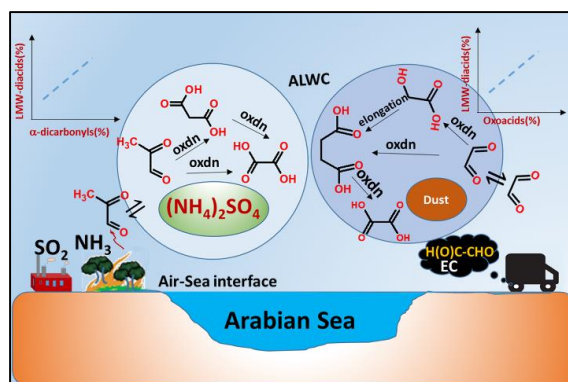
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Abstract

Oxalic acid and the related organics are ubiquitous water-soluble constituents of ambient organic aerosols (OA). Their molecular distributions and, relative abundances provide crucial insights on the secondary OA formation processes in the marine atmosphere and, are important for understanding the cloud-aerosol interactions. Here, we measured dicarboxylic acids, pyruvic acid, ω-oxocarboxylic acids and α-dicarbonyls alongside aerosol chemical composition in the South Asian outflow to the Arabian Sea. Oxalic acid (C₂) was the most abundant dicarboxylic acid followed by succinic (C₄) and malonic (C₃) acids; this distribution is typical of “less aged OA”. Oxalic acid levels correlated well with aerosol liquid water content (ALWC) and non-sea-salt (nss)-SO₄²⁻ (a tracer of coal combustion influence), suggesting an important aqueous phase oxidation of successive higher homologs. This was inferred based on the prevailing inverse relationships between the relative abundances of oxalic acid in total aliphatic dicarboxylic acids and those of malonic, succinic, glutaric, adipic, and azelaic acids. Likewise, the concentrations of glyoxal (Gly: a major compound emitted from vehicular exhaust) and glyoxylic acid (ωC₂) correlated strongly with C₄, nss-Ca²⁺ (dust tracer) and elemental carbon (EC: a combustion tracer from biomass/fossil-fuels). These observations suggest gas/particle partitioning and subsequent aqueous phase oxidation of Gly to ωC₂ and then elongation (oligomerization) to succinic acid associated with the mineral dust particles. This means relative abundances of anthropogenic SO₄²⁻ in the dust-laden air masses govern the secondary OA formation over the Arabian Sea.

Keywords: oxalic acid, glyoxylic acid, pyruvic acid, aerosol liquid water content, aerosol acidity, mineral dust.

TOC abstract



1. Introduction

The dicarboxylic acids and related compounds constitute a major fraction of atmospheric organic aerosols (OA). Their molecular distributions and relative abundances provide crucial insights related to the secondary organic aerosol (SOA) formation processes. Some evidence support that coatings of dicarboxylic acids with the soot particles (aka black or elemental carbon; EC) can significantly influence its hygroscopic properties, shape and density ([Xue et al., 2009](#)). Oxalic acid and their successive homologs are one such SOA and are well-known for their ubiquitous abundance in urban, rural and remote locations([Kawamura and Bikkina, 2016](#)). These organic compounds are, in general, either formed by the degradation of long-chain successive homologs (*e.g.* azelaic acid; ([Bikkina et al., 2014](#))) or by the secondary processes such as condensation of semi volatile organic compounds (SVOC) and their subsequent aqueous phase chemical reactions in clouds and/or aerosol water content ([Bikkina et al., 2017](#)).

Studies focusing on low molecular weight (LMW) dicarboxylic acids and related compounds in marine aerosols influenced by the continental outflows provide important insights related to aging of OA ([Yang et al., 2020b](#)). However, in comparison to land observations, studies from the marine regions are rather limited in the literature but those marine studies could provide detailed insights on the formation and evolution of OA during atmospheric transport ([McNeill, 2015](#)). For instance, the molecular distributions of dicarboxylic acids have been used to track the penetration of biomass burning plumes from the Indo-Gangetic Plain (IGP) across the higher Himalayan Mountains ([Cong et al., 2015](#)). Likewise, [Bikkina et al. \(2014\)](#) have documented the relative importance of marine biogenic emissions during a phytoplankton bloom in the western North Pacific in contributing to the atmospheric abundances of oxalic acid and their precursor organics.

Another study by [Hoque and Kawamura \(2016\)](#) have observed that the atmospheric levels of dicarboxylic acids over the Equatorial Pacific are somewhat influenced by the photochemical oxidation processes of marine derived organic compounds. [Kawamura and Sakaguchi \(1999\)](#) have proposed that photochemical oxidation processes occurring during transport largely dictate the atmospheric abundances of oxalic acid (a major SOA) and related polar compounds in marine aerosols influenced by the continental outflow from the East Asia to the western North Pacific. [Bikkina et al. \(2017\)](#) have highlighted the role of anthropogenic SO_4^{2-} in controlling the abundances of oxalic acid and precursors through aqueous phase chemical reactions during the impact of South Asian outflow. However, no such work has been carried out in the Arabian Sea so far that is dealing with the measurements of dicarboxylic acid and related polar compounds.

The northern Indian Ocean, comprised of the Arabian Sea on the west and the Bay of Bengal on the east, is strongly influenced by the long-range transport of haze pollution occurring over South Asia in winter season due to prevailing NE-monsoonal winds ([Lelieveld et al., 2001](#); [Gustafsson et al., 2009](#)). Although some studies have focused on the characterization of oxygenated OA from the tropical Indian Ocean ([Yang et al., 2020a](#); [Alang et al., 2022](#); [Hegde et al., 2022](#)), most of these are related to two decades old observations and, that too focusing on hazardous air pollutants such as polycyclic aromatic hydrocarbons (PAHs), lipids and sugar compounds ([Fu et al., 2011](#); [Fu et al., 2013](#)). However, studies measuring the dicarboxylic acids and related polar compounds in marine aerosols from the Indian Ocean are rather limited to constrain their likely sources and formation processes over the Arabian Sea. Therefore, the major goal of this study is to determine the concentration levels of major dicarboxylic acids, pyruvic acid, ω -oxocarboxylic acids and α -dicarbonyls in wintertime marine aerosols collected over the Arabian Sea during the impact of South Asian outflow. These results could provide necessary background information to interpret the molecular distributions and relative abundances upon examining the interrelationships with other chemical tracers of dust and pollution (*e.g.*, levoglucosan, anthropogenic SO_4^{2-} , elemental carbon; EC, *aka* soot) as well as stable carbon isotopic ratios of total carbon to understand the probable sources and formation pathways.

2. Materials and methods

2.1. Aerosol collection

Total suspended particulate matter (TSP) samples (N=17) were collected on prebaked (at 450°C for 6 h) Tissuquartz filters (dimensions: 8" ×10"; PALLFLEX®™) by using a high volume (TSP; Envirotech®™ model, APM 430) air sampler during a winter cruise in the Arabian Sea (6-24 December 2018). The Sagar Sampada (SS379) expedition started from Kochi on 6 December 2018 and ended at Okha port, Gujarat on 24 December 2018. The TSP air sampler was installed onboard on the bridge of ORV *Sagara Sampada* in the front side during the SS379 cruise (Fig. 1). Sufficient care has been taken to ensure contamination free sampling from ship's exhaust by collecting aerosols only when our research vessel is cruising at a speed of 8-10 knots per hour and relative wind is from the fore side. For more details about the cruise track and meteorology, reference were made to our earlier publications ([Bikkina et al., 2020](#); [Bikkina et al., 2021a](#); [Bikkina et al., 2022a](#); [Bikkina et al., 2022b](#)). After collection, aerosol filters were wrapped in prebaked Aluminum foils and stored at -20 °C. All the sample extraction, processing and analysis were performed in Prof. Kawamura's laboratory at Chubu University, Japan using the well-established analytical technique for dicarboxylic acids,

pyruvic acid and ω -oxocarboxylic acids and α -dicarbonyls ([Kawamura and Bikkina, 2016](#)). Briefly, the extraction, derivatization and analytical protocol are outlined here.

2.2. Analysis of dicarboxylic acids and related compounds

A portion of the TSP filter was extracted with ultra-pure deionized water (specific resistivity > 18.2 M Ω -cm) by ultrasonic agitation and passed through a Pasteur pipette packed with quartz wool. The extract, after adjusting the solution pH to 8.5-9.0, was concentrated to near dryness under vacuum by using a rotary evaporator and reacted with 60 μ l of 14% BF₃ in *n*-butanol (Supelco[®]™) using a hot water bath at 100 °C for 1 h. This step converts the dicarboxylic acids to their respective dibutyl esters and ω -oxocarboxylic acids and α -dicarbonyls into their hemiacetals. Soon after, these derivatives were back extracted into *n*-hexane and preconcentrated in small volumes (~500 μ l) using the rotary evaporator. This *n*-hexane concentrate was dried using N₂ blow down technique and made up to suitable volume in *n*-hexane. Approximately 2 μ l of the derivatized extract was injected onto a capillary gas chromatograph (GC, HP5890) equipped with split/splitless mode and connected inline to a flame ionization detector (FID).

The concentrations were obtained by comparing the relative peak areas of target analytes with the standards derivatized similarly like real aerosol samples. The recoveries obtained for major dicarboxylic acid standards via spiking on the blank filters and extracted like real aerosol samples were better than 90% except for oxalic acid, for which we obtained a recovery of 80%. We also used the concentrations of Levoglucosan (*Lev*), water-soluble inorganic ions, elemental carbon (EC), organic carbon (OC) and water-soluble OC (WSOC), stable carbon isotopic composition ($\delta^{13}\text{C}$) of total carbon (TC) from our earlier publications to strengthen our arguments related to different source influence over the Arabian Sea ([Bikkina et al., 2020](#); [Bikkina et al., 2022b](#)).

2.3. Air mass back trajectories and fire counts

To ascertain the influence of dust and probable source regions of the air parcels advected to the Arabian Sea during the SS379 cruise, we computed air mass back trajectories using the offline computer based Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT, version-4 ([Draxler et al., 2012](#); [Stein et al., 2015](#))) alongside the dust aerosol optical thickness retrievals at 550 nm from the Modern-Era Retrospective analysis for Research and Applications, Version 2 (MERRA-2) database, accessible through the Global Modeling and Assimilation Office (GAMO) web portal (Fig. 1). We used the archived reanalysis meteorological fields from the National Center for Environmental Prediction and National Center for Atmospheric Research (NCEP/NCAR) web portal as an input to the HYSPLIT model. Likewise, we also obtained the Moderate resolution imaging

spectroradiometer (MODIS) fire counts ([Justice et al., 2002](#)) during the study period to understand the relative importance of crop-residue burning and open fires over South Asia on the atmospheric abundances of dicarboxylic acids and related polar compounds.

2.4. Aerosol liquid water content (ALWC) and aerosol acidity

Several studies have suggested that the particulate bound water and associated aerosol acidity play an important role in the formation of various SOA ([Deshmukh et al., 2016](#); [Bikkina et al., 2017](#)). However, the relative influence and importance of these two variables are not clear for oceanic regions that are downwind of pollution sources in South Asia. At present, several studies in the literature basically use thermodynamic equilibrium partition models, ISORROPIA (<http://isorro피아.eas.gatech.edu>) and Extended Aerosol Inorganic ion Model: E-AIM (<http://www.aim.env.uea.ac.uk/aim/aim.php>), to predict the ALWC and aerosol acidity, respectively, based on the concentrations of water-soluble inorganic ions. The ISORROPIA-II module will only allow the input of (either in units of $\mu\text{g m}^{-3}$ or $\mu\text{mol m}^{-3}$) inorganic ionic species (Na^+ , SO_4^{2-} , NH_4^+ , NO_3^- , Cl^- , Ca^{2+} , K^+ , and Mg^{2+}) and does not have option to include certain organic acids (e.g., oxalic acid) like the E-AIM (either II or IV) ([Clegg et al., 1998](#)). In this study, we calculated the ALWC and aerosol acidity using the concentrations of water-soluble inorganic ions (Na^+ , SO_4^{2-} , NH_4^+ , Cl^- , NO_3^- , Ca^{2+} , K^+ , and Mg^{2+}) and meteorological fields (*i.e.*, relative humidity of 85% and temperature of 25 °C) in ISORROPIA-II, which was developed by [Fountoukis and Nenes \(2007\)](#). The choice of ISORROPIA over the E-AIM made here is just to consider the effect of crustal components on the ALWC and the formation of oxalic acid and related precursors. Numerous studies have pointed out the impact of long-range transport of dust over the Arabian Sea from the surrounding deserts in the Middle East and India ([Pease et al., 1998](#); [Tindale and Pease, 1999](#); [Kumar and Sarin, 2009](#); [Bikkina and Sarin, 2012](#)). Besides, recent studies on aerosol pH have compared these two models (ISORROPIA and E-AIM) across different urban sites in China and USA, and have shown only a rather small variability ([Battaglia Jr et al., 2019](#)). Therefore, aerosol pH value obtained from the ISORROPIA II module over the Arabian Sea is kind of representation for the alkaline nature of dust, which is also consistent with inferences drawn from other studies ([Rastogi and Sarin, 2006](#); [Kumar et al., 2008a](#)). We also attempted to use the E-AIM model for estimating the ALWC, however, it has no option to include the concentrations of major ionic species associated with mineral dust (Ca^{2+} , K^+ and Mg^{2+}). Since the dust is a perennial constituent over the Arabian Sea, the E-AIM could not be run for most of the samples unlike ISORROPIA-II due to the inherent discrepancy of charge balance between the cation and anion species from the SS379 data.

3. Results and discussion

3.1. Spatial variability of concentrations

The concentrations of dicarboxylic acids and related polar compounds in marine aerosols collected over the Arabian Sea exhibited a pronounced spatial variability (Fig. 2). A summary of the statistical description of their concentration range, median, mean, and standard deviation in marine aerosols from the Arabian Sea is provided in Table 1. Of these, some of the tracer compounds are useful for probing the anthropogenic influence and relative significance of aqueous phase chemical reactions to better constrain their formation processes in the continental outflows (Fig. 3). For instance, oxalic acid (C_2) and their successive homologs malonic (C_3) and succinic (C_4) acids are, in general, considered as photochemical degradation products of adipic (C_6), azelaic (C_9), phthalic (Ph), terephthalic (tPh), pyruvic (Pyr), and glyoxylic (ωC_2) acids, glyoxal (Gly) and methylglyoxal (MeGly) through the gas and aqueous phase oxidation pathways ([Kawamura et al., 1996](#); [Kawamura and Sakaguchi, 1999](#); [Altieri et al., 2006](#); [Carlton et al., 2006](#); [Carlton et al., 2007](#); [Altieri et al., 2008](#); [Ervens et al., 2008](#); [Tan et al., 2010](#); [Bikkina et al., 2017](#); [Bikkina et al., 2021b](#)).

We compared the concentrations of C_2 , C_6 , Ph, C_9 , Gly and MeGly in marine aerosols from the Arabian Sea with those reported over other marginal seas and open ocean waters (Fig. 4). The oxalic acid (C_2), a major ubiquitous SOA ([Bikkina et al., 2021b](#); [Shen et al., 2022](#)) and the most abundant dicarboxylic acid, usually formed by the oxidation of their successive homologs and related compounds formed by the α -dicarbonyls ([Carlton et al., 2006](#); [Carlton et al., 2007](#); [Ervens et al., 2008](#); [Tan et al., 2010](#)). The concentrations of oxalic acid over the Arabian Sea are lower as compared to those reported over the Bay of Bengal during the impact of IGP-outflow and the biomass burning influenced receptor sites in Mt. Tai, North China Plain ([Kawamura et al., 2013](#)) and Rondônia, Brazil ([Kundu et al., 2010](#)) (Fig. 4a). It is important to reiterate the relative difference in the source emissions of OA and dust between the two limbs of the northern Indian Ocean. The Arabian Sea is more prone to dust deposition as it is surrounded by the deserts of Middle East and India ([Pease et al., 1998](#); [Tindale and Pease, 1999](#); [Kumar et al., 2008a](#); [Kumar et al., 2008b](#)). In contrast, the Bay of Bengal is more susceptible to the biomass burning and fossil fuel combustion emissions. [Liu et al. \(2013\)](#) observed a slight decrease in SOA yield in the presence of mineral dust seed particles. Therefore, relatively low concentrations of oxalic acid over the Arabian Sea than the Bay of Bengal during the IGP-outflow could be due to suppressed oxidation of precursors to C_2 on mineral dust surface. However, the concentrations of oxalic acid over the Arabian Sea are somewhat higher than the Bay of Bengal during the impact of Southeast Asian outflow as well as those documented over the Pacific and Arctic Oceans. Likewise, the C_2 loadings over the Arabian Sea are comparable to another receptor site in Jeju Island

in South Korea influenced by mineral dust and pollutants in the East Asian outflow to the western North Pacific. Overall, the variations in the atmospheric abundances of C₂ are likely due to differences in their precursor levels and oxidation pathways.

We examined the relative concentration levels of other important precursor dicarboxylic acids (C₆, Ph, C₉) and α -dicarbonyls (Gly, MeGly) over the Arabian Sea and other marginal seas (Fig.4). Grosjean et al. (1994) observed the formation of C₆ and Ph by the oxidation of cyclic olefins and aromatic hydrocarbons, which are abundant in fossil fuel combustion exhaust from motor vehicles, with atmospheric NO and O₃ under sunlight conditions. Likewise, C₉ is a photochemical oxidation product of biogenic unsaturated fatty acids (*i.e.* originated from land plants or marine phytoplankton exudates) in ambient aerosols. The measurements carried out inside Chinese tunnels by Zhang et al. (2016) have provided insights about the predominance of Gly over MeGly. Recently, Zarzana et al. (2018) have conducted several laboratory test burns of different biomass types and observed that MeGly emissions are a 2-3 factor higher than Gly. We can use this information to understand the relative importance of various source-emissions to C₂ precursors over the Arabian Sea with that of other marginal seas and open ocean measurements elsewhere.

As analogous species to C₂, we compared the concentrations of related precursors (C₆, Ph, C₉, and α -dicarbonyls (Gly, MeGly)) in marine aerosols from the Arabian Sea with those collected over other marginal seas (Fig. 4b-f). If the C₂ was, indeed, formed by a photochemical oxidation of precursor species in the continental outflow from South Asia, then we would expect their atmospheric loadings like this major dicarboxylic acid. Except MeGly, the concentrations of possible precursor organics (C₆, Ph, C₉, and Gly) were higher over the Arabian Sea compared to those over the Bay of Bengal, Gosan, Chichijima, the Pacific and Arctic Oceans and Mt. Everest. However, the atmospheric loadings of all these C₂ precursors over the Arabian Sea were lower than those from the receptor sites influenced by biomass burning.

3.2. Molecular distributions

A comparative assessment of the most abundant LMW dicarboxylic acids (*aka.* LMW-diacids: C₂-C₄) is useful for obtaining the information related to extent of aging of OA in the continental outflows. Kawamura and Bikkina (2016) have reviewed the concentration levels of LMW-diacids in marine aerosols from different oceanic regions. One prominent finding from their study(Kawamura and Bikkina, 2016) is that C₄ levels often dominate after C₂ over neritic waters, whereas the concentrations of C₃ exceed that of C₄ over pelagic oceans(Kawamura and Sakaguchi, 1999; Wang et al., 2006; Hoque et al., 2015; Deshmukh et al., 2018; Hoque et al., 2020). Therefore, the spatial

variability of molecular distribution of LMW-diacids over the Arabian Sea provides clues related to “relatively fresh” or “aged” characteristics of continental OA in the South Asian outflow (Fig. 5a). Notably, C₂ was the most abundant LMW-diacid followed by C₄ and C₃ in all the TSP samples (except SS379#12) collected over the Arabian Sea, suggesting “relatively fresh nature” of these SOA. This result is also in sharp contrast to the molecular distributions of LMW-diacids documented over the Bay of Bengal (C₂ > C₃ > C₄) during the impact of South Asian outflow (27 December 2008-28 January 2009)([Bikkina et al., 2017](#)).

Despite being influenced by similar source-emissions (*i.e.* crop-residue/wood burning and fossil-fuel combustion), we observed contrasting patterns of second most abundant dicarboxylic acid (C₃ or C₄) in marine aerosols over the Arabian Sea (this study) and Bay of Bengal([Bikkina et al., 2017](#)) during winter season. This observation, thus, emphasizes a pronounced aging effect of marine OA over the Bay of Bengal than the Arabian Sea during the impact of South Asian outflow. C₂ is not only the dominant one in the LMW-diacids but also among the measured aliphatic C₂-C₁₂ dicarboxylic acids (Fig. 5a). In general, the molecular distributions of measured related compounds are characterized by a predominance of Pyr, Gly, or 3-oxopropanoic acid (ω C₃) followed by other successive homologs measured (Fig. 5b). However, in two samples (SS379#2, SS379#5), we found the predominance of 8-oxooctanoic acid (ω C₈). In all the samples, we observed that Gly concentrations exceeded that of MeGly in the marine aerosols collected over the Arabian Sea (Fig. 5c).

3.3. Interrelationships

The interrelationships among the major dicarboxylic acids provide valuable information related to their common source emissions or formation processes occurring during long-range transport in the South Asian outflow to the Arabian Sea. Given the number of samples (N =17), the correlation coefficient (*r*) value over 0.48 is significant and, hence, highlighted in bold font in Fig. 6. Among the aliphatic dicarboxylic acids, we found that C₂ strongly correlated with C₃ but not with the C₄. Likewise, we observed good correlations among C₄, C₅, and C₆. Besides, C₄ also exhibited significant correlation with C₉, a photochemical oxidation product of marine biogenic unsaturated fatty acids (such as oleic acid) emitted from the ocean surface.

Correlation analysis between the measured dicarboxylic acids and other continental tracers of mineral dust (water-soluble Ca²⁺), anthropogenic SO₄²⁻ (nss-SO₄²⁻), biomass burning (*Lev*), and combustion sources (EC) could further provide clues about their sources and formation pathways (Fig. 6). Therefore, we only investigated those correlations that are significant according to Pearson’s correlation table. It is interesting to note that Gly correlated strongly with ω C₂ and C₄, but not with

Pyr. Besides this, Gly moderately correlated with nss-Ca^{2+} (a proxy for mineral dust (Rastogi and Sarin, 2006)) but strongly correlated with EC, OC and WSOC (Fig. 6). However, such significant correlation was absent between C_4 and *Lev* (*i.e.*, a biomass burning tracer; (Simoneit, 2002)). In contrast, both the C_2 and C_3 levels were moderately correlated with that of MeGly over the Arabian Sea. Likewise, these two LMW-dicarboxylic acids were not correlated with nss-Ca^{2+} , *Lev* and nss-K^+ (Fig. 6). These interrelationships provide intricate details about the formation mechanism of C_2 and other major homologs.

The Arabian Sea is more prone to the dust influence from surrounding deserts in the Middle East, Pakistan and India compared to the adjacent marine basin, the Bay of Bengal, which is also influenced by the continental outflow from South Asia during winter (Pease et al., 1998; Tindale and Pease, 1999; Kumar et al., 2008b; Badarinath et al., 2010). Some studies focusing on the size-distribution of dust-laden aerosols have documented that C_2 peaked with nss-Ca^{2+} concentrations (Wang et al., 2015; Deshmukh et al., 2017), suggesting heterogeneous formation of this LMW-dicarboxylic acid on the mineral aerosol surface. Ponczek et al. (2019) studied the photooxidation process of dicarboxylic acid coated on mineral dust particles eventually resulted in the formation of multifunctional derivatives. In this context, the observed linear relationships of Gly and C_4 with nss-Ca^{2+} (Fig. 6) further suggest a plausible condensation of former SVOC onto the airborne mineral dust during transport and its subsequent oligomerization (chain-elongation) to C_4 .

We also observed a strong linear relationship of Gly with EC (Fig. 6) but not with the *Lev* and nss-K^+ in marine aerosols collected over the Arabian Sea during winter (Fig. 6). This means Gly might have originated from the fossil-fuel combustion sources. Moreover, the lack of correlation between Gly and nss-SO_4^{2-} (*i.e.*, an oxidation product of SO_2 emitted from the coal-fired thermal power plants) further corroborates the importance of vehicular emissions. The VOC measurements carried out in the road tunnels of China and USA have revealed the importance of vehicular emissions as a source of Gly and MeGly. Of these, the emission factors of α -dicarbonyls determined from the Chinese road tunnel experiments by Zhang et al. (2016) further revealed a major share of Gly ($1.18 \pm 0.43 \text{ mg km}^{-1} \text{ vehicle}^{-1}$) over MeGly ($0.52 \text{ mg km}^{-1} \text{ vehicle}^{-1}$) from the vehicular emissions. Taken these observations altogether, we speculate that Gly mostly originated from vehicular emissions in the South Asian outflow to the Arabian Sea. It is uncertain at this moment what factors would lead to such observed strong correlations of Gly with glyoxylic acid (Fig. 6) and C_4 (Fig. 6), but not with the Pyr. We speculate here that ALWC might play a role in the formation of dicarboxylic acids and related water-soluble compounds in dust-laden marine aerosols.

3.4. Diagnostic ratios

The spatial variability in the concentration ratios of certain dicarboxylic acids and related polar compounds (C_2/C_4 , C_3/C_4 , M/F, C_2/Pyr , $C_2/\omega C_2$, C_2/Gly , C_2/MeGly , and C_6/C_9 ; Fig. 7a-h) alongside their relationships with other chemical tracers (*e.g.* *Lev*, EC, nss-Ca^{2+} and nss-K^+) could provide insights on their formation pathways (Pavuluri et al., 2015). For instance, the photochemical oxidation experiments of C_3 , C_4 , Gly, MeGly, Pyr, and ωC_2 have yielded C_2 (Carlton et al., 2006; Carlton et al., 2007; Yang et al., 2008). We observed similar spatial variability of C_2/C_4 , C_3/C_4 , M/F, $C_2/\omega C_2$, C_2/Gly over the Arabian Sea in marine aerosols collected over 10-20°N compared to those sampled over 20-25°N and 5-10°N (Fig. 7a-c, Fig. 7e-f). However, C_2/Pyr has exhibited distinct spatial variability with the persistent high ratios over 5-10°N (Fig. 7d). As Pyr is an efficient tracer for the oxidation processes involving biogenic VOCs (*e.g.*, isoprene) in clouds and wet aerosols (Carlton et al., 2006), relatively high ratios of C_2/Pyr towards the southern Arabian Sea are likely because of the increase in ambient temperature and relative humidity. In contrast, we observed distinct spatial variability for the diagnostic ratios of C_2/MeGly (Fig. 7g) and C_6/C_9 (Fig. 7h) with high ratios observed only at two discrete latitudes, 20°N and 10-12°N. MeGly is also a biogenic VOC (*e.g.*, isoprene emitted from productive oceans such as Arabian Sea) oxidation product in the marine atmosphere (Bikkina et al., 2014). Likewise, C_9 is a photochemical oxidation product of biogenic unsaturated fatty acid emitted from the ocean surface during high biological activity (Bikkina et al., 2014). Studies have shown that C_6 is the photochemical oxidation product of cyclic olefins emitted from the vehicular exhaust as well as by the degradation of C_9 (Kawamura et al., 1996; Kawamura and Sakaguchi, 1999). Therefore, the spatial distributions of these two ratios, C_2/MeGly and C_6/C_9 are likely similar because of their biogenic origin from the surface ocean (Fig. 7).

3.5. Role of aerosol liquid water content (ALWC) and anthropogenic SO_4^{2-}

Many studies have highlighted the importance of particle-bound water in ambient aerosols in the formation of C_2 and its precursor organics (McNeill, 2015). There is some evidence in the literature describing that heterogeneous uptake of acidic anthropogenic SO_4^{2-} could increase the water content in aerosols, which enables partitioning of SVOC (*e.g.* Gly, MeGly) and subsequent oxidation processes contribute to the formation of C_2 and their successive homologs (Bikkina et al., 2017). We, therefore, looked out for any relationship existed between the dicarboxylic acids/precursors measured and ALWC calculated.

Of all the measured dicarboxylic acids and related compounds, C_2 correlated well with both the ALWC (Fig. 8a) and nss-SO_4^{2-} (Fig. 8b) in marine aerosols collected over the Arabian Sea during

our winter cruise. This observation essentially highlights the aqueous phase oxidation of precursor organics to C_2 in the acidic SO_4^{2-} containing marine aerosols. Here, the slope of linear regression between C_2 and $nss-SO_4^{2-}$ is comparable to previously reported values over the Bay of Bengal during the impact of IGP and South East Asian outflows during winter (Bikkina et al., 2017). Furthermore, the average molar ratio of $C_2/nss-SO_4^{2-}$ across global oceans including the present study is provided in Table 2. Yu et al. (2005) argued, based on the comparison of such relationships from the ambient aerosols collected in East Asia and the East Asian outflow to the western North Pacific, that C_2 is mostly formed through the *in-cloud* oxidation pathways or in the wetted aerosols (particles with high ALWC). Likewise, Sorooshian et al. (2006) observed strong correlation between oxalate and SO_4^{2-} in cloud water influenced by the power plant emissions over northern America. Therefore, the linear relationships observed between C_2 and $nss-SO_4^{2-}$ further indicate their formation through aqueous phase oxidation processes (*i.e.*, as we sampled 10 m above sea level because of which *in-cloud* role has been ruled out here).

Away from the source-emissions and secondary formation processes over the continents, the dilution of air parcels would similarly affect both these chemical species during transport. $nss-SO_4^{2-}$ in ambient aerosols from South Asia largely come from the oxidation of SO_2 emitted from coal-fired thermal power plants. Likewise, C_2 is a photochemical oxidation product of most of the organic compounds of anthropogenic origin (Kawamura et al., 1996; Kawamura and Bikkina, 2016). Although some studies reported higher abundances of C_2 from biomass burning, these aerosols were mostly collected near the receptor sites. Recent time-series SOA experiments focusing on the isoprene oxidation revealed the formation of C_2 within a few minutes, even in the gas phase (Bikkina et al., 2021b). This means C_2 emissions from the biomass burning source regions are also kind of oxidation products formed within the fire premises. Likewise, Huang and Yu (2007) observed rather small contributions of C_2 from vehicular emissions sampled inside road tunnels in China. All these results further prompting us to conclude that high abundances of C_2 in marine aerosols over the Arabian Sea during winter season are mostly of secondary origin.

Some studies focusing on the size-distribution of coastal and continental aerosols have documented the predominant fine mode nature of both C_2 and $nss-SO_4^{2-}$ in aerosols (Crahan et al., 2004). Therefore, gravitational settling might not be the influential factor here. Instead, their principal removal mechanism in the continental outflows is perhaps due to the wet scavenging (Yu et al., 2005). In addition, both these compounds are highly hygroscopic and thus easily partition into clouds and/or adsorb onto the surface of deliquescent aerosols during long-range transport. Apart from the droplet mode existence (0.26-0.44 μm), Crahan et al. (2004) observed high concentrations of SO_4^{2-} and C_2 in

cloud water than the below cloud aerosols. Another possibility for the observed linear relationship could be the overall control of nss-SO_4^{2-} levels on the ALWC in marine aerosols over the Arabian Sea. In such scenario, the ALWC would provide a medium for precursor homologs of C_2 and other SVOCs to partition in the liquid phase and, thus, facilitate further aqueous oxidation process.

Earlier studies from the western North Pacific have highlighted that concentrations of C_9 are substantially high due to concurrent biological activity in the surface waters ([Bikkina et al., 2014](#)). Since the Arabian Sea is one of the globally recognized productive marine basin ([Madhupratap et al., 1996](#)), comparable levels of C_9 with C_4 and C_3 in the ambient air during our winter cruise further revealed the significance of oceanic source in addition to the continental impact from South Asia. Moreover, the photochemical oxidation experiments involving the C_9 initially resulted in the formation of successive lower homologs from C_2 to C_6 ([Yang et al., 2008](#)). Furthermore, photochemical oxidation of C_3 , C_4 , C_5 and C_6 have resulted in the formation of C_2 ([Yang et al., 2008](#)). If the C_2 is formed from these reactants by the oxidation reactions in the aerosol water, we would expect an inverse relationship between precursors (C_3 , C_4 , C_5 , C_6 and C_9) and product (C_2) over the Arabian Sea. However, to avoid variability introduced by the other source-emissions and formation pathways (*e.g.*, Gly, MeGly, Pyr and ωC_2 can produce C_2 in the liquid water), we first normalized the concentrations of these precursor organics and C_2 with the total mass loading of aliphatic dicarboxylic acids ($\Sigma\text{C}_2\text{-C}_{12}$) before investigating their interrelationships. We observed a significant inverse linear relationships between these normalized concentration ratios of precursor organics and the C_2 (Fig. 8c-d). Overall, it is likely that observed linear relationship between C_2 and nss-SO_4^{2-} in marine aerosols are likely due to the combined role of aqueous phase oxidation processes involving successive homologs of C_2 and related SVOCs (Gly and ωC_2) over the Arabian Sea during winter season.

3.6. Relative abundances

The relative abundances of C_2 , the dominant SOA measured here, accounted for $\sim 0.21 - 5.6\%$ of WSOC over the Arabian Sea (av. 1.6%) during the impact of South Asian outflow. This is comparable to the relative abundance of C_2 in WSOC from the IGP-outflow samples over the Bay of Bengal (range: 1.0-3.0%; av. 1.9%; $P > 0.05$). However, C_2 levels over the Arabian Sea ($15\text{-}352 \text{ ng m}^{-3}$) were lower than the Bay of Bengal ($104\text{-}661 \text{ ng m}^{-3}$) during the impact of wintertime IGP-outflow ([Bikkina et al. \(2017\)](#)). Another striking difference in the chemical composition of marine aerosols between these two marine basins during the IGP-outflow is found in the atmospheric abundances of nss-SO_4^{2-} (Arabian Sea: $3\text{-}16 \text{ } \mu\text{g m}^{-3}$; Bay of Bengal: $2\text{-}28 \text{ } \mu\text{g m}^{-3}$) and nss-Ca^{2+} (Arabian Sea: $0.5\text{-}3.5 \text{ } \mu\text{g m}^{-3}$; Bay of Bengal ([Bikkina and Sarin, 2012](#)): $0.02\text{-}0.07 \text{ } \mu\text{g m}^{-3}$). Earlier studies have highlighted

that the Arabian Sea is strongly influenced by the mineral dust, while the impact of continental pollution is more severe over the Bay of Bengal ([Kumar et al., 2008b](#); [Bikkina and Sarin, 2012](#); [Yang et al., 2020b](#)). Besides, [Bikkina et al. \(2017\)](#) have highlighted the importance of anthropogenic SO_4^{2-} in the formation of C_2 over the Bay of Bengal.

We observed similar relative abundances of C_2 in WSOC over these two marine basins despite significant differences in the molar ratios of $\text{C}_2/\text{nss-SO}_4^{2-}$ between the dust-laden air masses from the Arabian Sea (0.018) and the pollution-affected marine aerosols from the Bay of Bengal (~ 0.006). This observation essentially highlights the role of dust as a surrogate for the oxidation of precursor organics to produce C_2 over the Arabian Sea. However, relatively low concentrations of anthropogenic SO_4^{2-} over the Arabian Sea further explain the concordant C_2 loadings and, therefore, highlight the role of aerosol acidity driven ALWC for the SOA formation. Our estimate suggests that the dicarboxylic acids, oxocarboxylic acids and α -dicarbonyls contribute to 1.2-21%, 0.3-4.5% and 0.02-0.5% of WSOC, respectively, over the Arabian Sea during the impact of South Asian outflow. Likewise, these three compound classes account for 0.7-3.5%, 0.1-2.1%, and 0.01-0.06% of TC, respectively. We also estimated other statistical measures of the relative abundances of these three classes in WSOC, OC and TC (Table 3).

We observed a strong positive linear correlation between the relative abundance of LMW-diacids in WSOC with that of α -dicarbonyls (Fig. 9a). This feature indicates that α -dicarbonyls in the South Asian outflow to the Arabian Sea during winter season might have contributed to the formation of LMW-diacids. Several laboratory studies have observed that Gly and MeGly easily partition into the atmospheric particulate bound/cloud water during the long-range atmospheric transport ([Lim et al., 2013](#)). Besides, the correlation analysis between the water-soluble organics and continental chemical tracers (EC, nss-SO_4^{2-} , nss-Ca^{2+} and *Lev*) from the cruise indicated that Gly comes from vehicular exhaust and MeGly from the biomass burning in the continental outflow to the Arabian Sea. Additionally, strong positive linear relationships of C_2 with ALWC and nss-SO_4^{2-} alongside their interrelationship with other LMW-diacids, ωC_2 and α -dicarbonyls, altogether emphasize the significance of aqueous phase chemical reactions in the marine atmosphere.

Another important feature of the data is related to the persistent strong correlation between the equivalent carbon percentage contribution of oxoacids and the LMW-diacids in WSOC (Fig. 9b). However, this relationship is relatively weak compared to that evident between α -dicarbonyls/WSOC(%) and LMW-diacids/WSOC(%). [Kawamura et al. \(1996\)](#) have postulated that the relationship between oxoacids and LMW-diacids contribution to WSOC in marine aerosols are

likely because of their product to precursor formation by the ambient photochemical processes. However, the sources of oxoacids in marine aerosols are the photochemical oxidation processes involving biogenic unsaturated fatty acids either of continental or marine origin (Kawamura and Gagosian, 1987; Stephanou and Stratigakis, 1993). More recently, Li et al. (2022) have also suggested based on the molecular distributions of diacids and precursors as well as their stable carbon isotopic composition data that these intermediate oxoacids of unsaturated fatty acid origin could further oxidize to form straight chain dicarboxylic acids. Therefore, observed positive linear relationship between the percentage relative abundance of oxoacids and LMW-diacids in WSOC, thus, highlight the possible oxidation route/conversion of precursor oxo- intermediates to straight chain dicarboxylic acids in the dust laden marine aerosols from the Arabian Sea.

3.7. Formation pathways

Given the presence of mineral dust and ALWC together with the prevailing linear relationship of Gly with EC, ωC_2 , succinic and glutaric acids as well as the linear relationship of MeGly with Pyr overall indicate the formation of oxalic acid precursors from the fossil-fuel combustion and biomass burning sources in the South Asian outflow. Besides, the inverse relationships observed between relative abundance of oxalic acid with these precursor(s) further confirm oxidative degradation in the ALWC. Therefore, the key reaction pathways in the aqueous phase have been illustrated in Fig. 10 based on the literature review that support our field results from the Arabian Sea during the impact of South Asian outflow.

Glyoxal (Gly) is an abundant α -dicarbonyl in the continental outflows, typically produced by the oxidation of aromatic hydrocarbons in the vehicular exhaust over urban regions. Here, we observed rather high concentrations of Gly over the Arabian Sea during SS379 cruise conducted in winter season. Besides, the prominent linear relationships of Gly with EC but not with nss-SO_4^{2-} clearly emphasize their origin from automobile emissions in the South Asian outflow. Because of the high Henry's law constant ($H = 3 \times 10^5 \text{ M atm}^{-1}$ at 25°C ; (Tan et al., 2010)) and fast uptake rate into the atmospheric liquid droplets, the concentration of Gly is usually found to be high in cloud/wet aerosols ($\sim 10\text{-}100 \mu\text{M}$). Consequently, Gly can easily partition into the ALWC and undergoes oxidation to form glyoxylic acid, oxalic acid, malonic acid and succinic acid (Fig. 8; (Tan et al., 2009; Lee et al., 2011)).

Gly in the ALWC mostly exist as monomeric hydrated form (Fratzke and Reilly, 1986b, a), which in turn react with OH radical to form a glyoxyl alkyl radical intermediate (I). This step involves the abstraction of hydrogen atom by the OH radical from one of the carbon centre of geminal diol (Buxton et al., 1997). Furthermore, Schaefer et al. (2015) have observed that this alkyl radical could form an

adduct with dissolved oxygen to yield an alkylperoxy radical intermediate (II), which undergoes a rearrangement to yield glyoxylic acid (Fig. 10). [Lim et al. \(2013\)](#) have proposed similar reaction pathways for the formation of oxalic acid and pyruvic acid from glyoxylic acid and methylglyoxal, respectively (Fig. 10). However, pyruvic acid in the atmospheric aqueous phase mostly exists in the keto form (35%), which is in equilibrium with its hydrated form (~65%). The carbonyl functional group in the keto form of pyruvic acid enables this molecule to become receptive for ambient light in the near UV region ($\lambda \sim 320$ nm) and, hence, moves to higher energy states (n, π^*) ([Reed Harris et al., 2014](#)). Besides, this excited pyruvic acid molecule could abstract hydrogen from one of the O-H bonds of geminal diol followed by decarboxylation (*i.e.*, expel CO_2) and disproportionation (*i.e.*, H shift) to yield acetic and lactic acids in wet aerosols at the air-sea interface ([Griffith et al., 2013](#); [Rapf et al., 2017](#); [Kappes et al., 2021](#)) (Fig. 10). The acetic acid could further react with OH radical to produce alkyl radical, which in turn undergoes self-recombination reactions with another alkyl radical to form succinic acid (Fig. 10).

4. Summary and Conclusions

This study examines the molecular distributions and relative abundances of some major dicarboxylic acids, oxocarboxylic acids and α -dicarbonyls in marine aerosols collected over the Arabian Sea during the impact of South Asian outflow. The molecular distributions of dicarboxylic acids in the marine aerosols were characterized by the predominance of C_2 followed by C_4 and C_3 acids and, then by C_9 acid. Significant inverse correlations observed between the relative abundances of C_2 in total aliphatic dicarboxylic acids and those of C_3 , C_4 , C_5 , C_6 and C_9 altogether emphasize a plausible photochemical degradation of these precursor compounds to C_2 . Furthermore, we observed significant linear relationships of the loadings of C_2 with those of nss-SO_4^{2-} and ALWC over the Arabian Sea, suggesting the important aqueous phase reactions in the SOA production. We also found the predominance of Gly over MeGly, and its correlation with EC in the marine aerosols, further suggesting the significant vehicular emissions as a major source of this α -dicarbonyl over the Arabian Sea. However, this observation from the Arabian Sea is in sharp contrast to that documented from the Bay of Bengal during the impact of South Asian outflow (relatively high concentrations of MeGly from the biomass burning ([Bikkina et al., 2017](#))). Besides, Gly levels over the Arabian Sea significantly correlated with those of nss-Ca^{2+} (*i.e.*, a tracer of mineral dust) and glyoxylic acid (ωC_2), a highly water-soluble species usually abundant in the atmospheric aqueous phases, suggesting the partition of Gly into the aqueous phase and further oxidation to form ωC_2 . Overall, our results underscore the importance of deliquescent mineral dust and the associated anthropogenic SO_4^{2-} role in the production

of C₂ and its precursors from the aqueous phase reactions of Gly and C₉ over the Arabian Sea during the South Asian outflow.

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References

- Alang, A.K., Aggarwal, S.G., Singh, K., Soni, D., Kawamura, K., Water-soluble dicarboxylic acids, oxoacids and α -dicarbonyls in the tropical aerosols in coastal megacity Mumbai: molecular characteristics and formation processes, *Journal of Atmospheric Chemistry*(2022), pp. 1-19.
- Altieri, K., Carlton, A.G., Lim, H.-J., Turpin, B.J., Seitzinger, S.P., Evidence for Oligomer Formation in Clouds: Reactions of Isoprene Oxidation Products, *Environmental Science & Technology* **40**(2006), pp. 4956-4960.
- Altieri, K.E. *et al.*, Oligomers formed through in-cloud methylglyoxal reactions: Chemical composition, properties, and mechanisms investigated by ultra-high resolution FT-ICR mass spectrometry, *Atmospheric Environment* **42**(2008), pp. 1476-1490.
- Badarinath, K.V.S. *et al.*, Long-range transport of dust aerosols over the Arabian Sea and Indian region — A case study using satellite data and ground-based measurements, *Global and Planetary Change* **72**(2010), pp. 164-181.
- Battaglia Jr, M.A., Weber, R.J., Nenes, A., Hennigan, C.J., Effects of water-soluble organic carbon on aerosol pH, *Atmos. Chem. Phys.* **19**(2019), pp. 14607-14620.
- Bikkina, P., Bikkina, S., Kawamura, K., Tracing the biomass burning emissions over the Arabian Sea in winter season: Implications from the molecular distributions and relative abundances of sugar compounds, *Science of The Total Environment* **848**(2022a), p. 157643.
- Bikkina, P., Bikkina, S., Kawamura, K., Sarma, V.V.S.S., Deshmukh, D.K., Unraveling the sources of atmospheric organic aerosols over the Arabian Sea: Insights from the stable carbon and nitrogen isotopic composition, *Science of The Total Environment* **827**(2022b), p. 154260.
- Bikkina, P., Sarma, V.V.S.S., Kawamura, K., Bikkina, S., Dry-deposition of inorganic and organic nitrogen aerosols to the Arabian Sea: Sources, transport and biogeochemical significance in surface waters, *Marine Chemistry* **231**(2021a), p. 103938.

- Bikkina, P. *et al.*, Chemical characterization of wintertime marine aerosols over the Arabian Sea: Impact of marine sources and long-range transport, *Atmospheric Environment*(2020), p. 117749.
- Bikkina, S., Kawamura, K., Miyazaki, Y., Fu, P., High abundances of oxalic, azelaic, and glyoxylic acids and methylglyoxal in the open ocean with high biological activity: Implication for secondary OA formation from isoprene, *Geophysical Research Letters* **41**(2014), pp. 3649-3657.
- Bikkina, S., Kawamura, K., Sakamoto, Y., Hirokawa, J., Low molecular weight dicarboxylic acids, oxocarboxylic acids and α -dicarbonyls as ozonolysis products of isoprene: Implication for the gaseous-phase formation of secondary organic aerosols, *Science of The Total Environment* **769**(2021b), p. 144472.
- Bikkina, S., Kawamura, K., Sarin, M., Secondary organic aerosol formation over Coastal Ocean: Inferences from atmospheric water-soluble low molecular weight organic compounds, *Environmental Science & Technology* **51**(2017), pp. 4347–4357.
- Bikkina, S., Sarin, M., Atmospheric pathways of phosphorous to the Bay of Bengal: contribution from anthropogenic sources and mineral dust, *Tellus B* **64**(2012).
- Buxton, G.V., Malone, T.N., Salmon, G.A., Oxidation of glyoxal initiated by OH in oxygenated aqueous solution, *Journal of the Chemical Society, Faraday Transactions* **93**(1997), pp. 2889-2891.
- Carlton, A.G. *et al.*, Atmospheric oxalic acid and SOA production from glyoxal: Results of aqueous photooxidation experiments, *Atmospheric Environment* **41**(2007), pp. 7588-7602.
- Carlton, A.G., Turpin, B.J., Lim, H.-J., Altieri, K.E., Seitzinger, S., Link between isoprene and secondary organic aerosol (SOA): Pyruvic acid oxidation yields low volatility organic acids in clouds, *Geophysical Research Letters* **33**(2006).
- Clegg, S.L., Brimblecombe, P., Wexler, A.S., Thermodynamic Model of the System $H^+ - NH_4^+ - SO_4^{2-} - NO_3^- - H_2O$ at Tropospheric Temperatures, *The Journal of Physical Chemistry A* **102**(1998), pp. 2137-2154.
- Cong, Z., Kawamura, K., Kang, S., Fu, P., Penetration of biomass-burning emissions from South Asia through the Himalayas: new insights from atmospheric organic acids, *Scientific reports* **5**(2015), p. doi:10.1038/srep09580.
- Crahan, K.K., Hegg, D., Covert, D.S., Jonsson, H., An exploration of aqueous oxalic acid production in the coastal marine atmosphere, *Atmospheric Environment* **38**(2004), pp. 3757-3764.
- Deshmukh, D.K., Kawamura, K., Deb, M.K., Boreddy, S.K.R., Sources and formation processes of water-soluble dicarboxylic acids, ω -oxocarboxylic acids, α -dicarbonyls, and major ions in summer aerosols from eastern central India, *Journal of Geophysical Research: Atmospheres* **122**(2017), pp. 3630-3652.
- Deshmukh, D.K., Kawamura, K., Lazaar, M., Kunwar, B., Boreddy, S.K.R., Dicarboxylic acids, oxoacids, benzoic acid, α -dicarbonyls, WSOC, OC, and ions in spring aerosols from Okinawa Island in the western North Pacific Rim: size distributions and formation processes, *Atmos. Chem. Phys.* **16**(2016), pp. 5263-5282.

- Deshmukh, D.K., Mozammel Haque, M., Kawamura, K., Kim, Y., Dicarboxylic acids, oxocarboxylic acids and α -dicarbonyls in fine aerosols over central Alaska: Implications for sources and atmospheric processes, *Atmospheric Research* **202**(2018), pp. 128-139.
- Draxler, R., Stunder, B., Rolph, G., Stein, A., Taylor, A., HYSPLIT 4 User Guide Overview. (2012).
- Ervens, B. *et al.*, Secondary organic aerosol yields from cloud-processing of isoprene oxidation products, *Geophysical Research Letters* **35**(2008).
- Fountoukis, C., Nenes, A., ISORROPIA II: a computationally efficient thermodynamic equilibrium model for K⁺-Ca²⁺-Mg²⁺-NH₄⁺-Na⁺-SO₄²⁻-NO₃⁻-Cl⁻-H₂O aerosols, *Atmos. Chem. Phys.* **7**(2007), pp. 4639-4659.
- Fratzke, A.R., Reilly, P.J., Kinetic analysis of the disproportionation of aqueous glyoxal, *International Journal of Chemical Kinetics* **18**(1986a), pp. 757-773.
- Fratzke, A.R., Reilly, P.J., Thermodynamic and kinetic analysis of the dimerization of aqueous glyoxal, *International Journal of Chemical Kinetics* **18**(1986b), pp. 775-789.
- Fu, P., Kawamura, K., Miura, K., Molecular characterization of marine organic aerosols collected during a round-the-world cruise, *Journal of Geophysical Research: Atmospheres* **116**(2011), p. doi: 10.1029/2011JD015604.
- Fu, P., Kawamura, K., Usukura, K., Miura, K., Dicarboxylic acids, ketocarboxylic acids and glyoxal in the marine aerosols collected during a round-the-world cruise, *Marine Chemistry* **148**(2013), pp. 22-32.
- Griffith, E.C., Carpenter, B.K., Shoemaker, R.K., Vaida, V., Photochemistry of aqueous pyruvic acid, *Proceedings of the National Academy of Sciences* **110**(2013), pp. 11714-11719.
- Grosjean, D., Williams, E.L., Grosjean, E., Novakov, T., Evolved Gas Analysis of Secondary Organic Aerosols, *Aerosol Science and Technology* **21**(1994), pp. 306-324.
- Gustafsson, Ö. *et al.*, Brown clouds over South Asia: biomass or fossil fuel combustion?, *Science* **323**(2009), pp. 495-498.
- Hegde, P., Boreddy, S.K.R., Aswini, A.R., Aryasree, S., Influence of south Asian outflow on secondary organic aerosol formation over the Indian Ocean: Inferences from water-soluble low molecular weight dicarboxylic acids and related organic compounds during ICARB 2018 experiment, *Marine Chemistry* **239**(2022), p. 104071.
- Hoque, M., Kawamura, K., Seki, O., Hoshi, N., Spatial distributions of dicarboxylic acids, ω -oxoacids, pyruvic acid and α -dicarbonyls in the remote marine aerosols over the North Pacific, *Marine Chemistry* **172**(2015), pp. 1-11.
- Hoque, M.M.M., Kawamura, K., Longitudinal distributions of dicarboxylic acids, ω -oxoacids, pyruvic acid, α -dicarbonyls, and fatty acids in the marine aerosols from the central Pacific including equatorial upwelling, *Global Biogeochemical Cycles* **30**(2016), pp. 534-548.
- Hoque, M.M.M. *et al.*, Molecular characteristics of water-soluble dicarboxylic acids, ω -oxocarboxylic acids, pyruvic acid and α -dicarbonyls in the aerosols from the eastern North Pacific, *Marine Chemistry* **224**(2020), p. 103812.

- Huang, X.-F., Yu, J.Z., Is vehicle exhaust a significant primary source of oxalic acid in ambient aerosols?, *Geophysical Research Letters* **34**(2007).
- Justice, C.O. *et al.*, The MODIS fire products, *Remote Sensing of Environment* **83**(2002), pp. 244-262.
- Kappes, K.J. *et al.*, Chemistry and Photochemistry of Pyruvic Acid at the Air–Water Interface, *The Journal of Physical Chemistry A* **125**(2021), pp. 1036-1049.
- Kawamura, K., Bikkina, S., A review of dicarboxylic acids and related compounds in atmospheric aerosols: Molecular distributions, sources and transformation, *Atmospheric Research* **170**(2016), pp. 140-160.
- Kawamura, K., Gagosian, R., Implications of ω -oxocarboxylic acids in the remote marine atmosphere for photo-oxidation of unsaturated fatty acids, *Nature* **325**(1987), p. 330.
- Kawamura, K., Kasukabe, H., Barrie, L.A., Source and reaction pathways of dicarboxylic acids, ketoacids and dicarbonyls in arctic aerosols: One year of observations, *Atmospheric Environment* **30**(1996), pp. 1709-1722.
- Kawamura, K., Sakaguchi, F., Molecular distributions of water soluble dicarboxylic acids in marine aerosols over the Pacific Ocean including tropics, *Journal of Geophysical Research: Atmospheres* **104**(1999), pp. 3501-3509.
- Kawamura, K. *et al.*, High abundances of water-soluble dicarboxylic acids, ketocarboxylic acids and α -dicarbonyls in the mountaintop aerosols over the North China Plain during wheat burning season, *Atmos. Chem. Phys.* **13**(2013), pp. 8285-8302.
- Kumar, A., Sarin, M.M., Mineral aerosols from western India: Temporal variability of coarse and fine atmospheric dust and elemental characteristics, *Atmospheric Environment* **43**(2009), pp. 4005-4013.
- Kumar, A., Sarin, M.M., Sudheer, A.K., Mineral and anthropogenic aerosols in Arabian Sea–atmospheric boundary layer: Sources and spatial variability, *Atmospheric Environment* **42**(2008a), pp. 5169-5181.
- Kumar, A., Sudheer, A., Sarin, M., Chemical characteristics of aerosols in MABL of Bay of Bengal and Arabian Sea during spring inter-monsoon: a comparative study, *Journal of Earth System Science* **117**(2008b), pp. 325-332.
- Lee, A.K.Y., Zhao, R., Gao, S.S., Abbatt, J.P.D., Aqueous-Phase OH Oxidation of Glyoxal: Application of a Novel Analytical Approach Employing Aerosol Mass Spectrometry and Complementary Off-Line Techniques, *The Journal of Physical Chemistry A* **115**(2011), pp. 10517-10526.
- Lelieveld, J. *et al.*, The Indian Ocean Experiment: Widespread Air Pollution from South and Southeast Asia, *Science* **291**(2001), pp. 1031-1036.
- Li, P. *et al.*, Year-round observations of stable carbon isotopic composition of carboxylic acids, oxoacids and α -Dicarbonyls in fine aerosols at Tianjin, North China: Implications for origins and aging, *Science of The Total Environment* **834**(2022), p. 155385.

- Lim, Y.B., Tan, Y., Turpin, B.J., Chemical insights, explicit chemistry, and yields of secondary organic aerosol from OH radical oxidation of methylglyoxal and glyoxal in the aqueous phase, *Atmos. Chem. Phys.* **13**(2013), pp. 8651-8667.
- Liu, C. *et al.*, Effect of mineral dust on secondary organic aerosol yield and aerosol size in α -pinene/NO_x photo-oxidation, *Atmospheric Environment* **77**(2013), pp. 781-789.
- Madhupratap, M. *et al.*, Mechanism of the biological response to winter cooling in the northeastern Arabian Sea, *Nature* **384**(1996), pp. 549-552.
- McNeill, V.F., Aqueous organic chemistry in the atmosphere: Sources and chemical processing of organic aerosols. ACS Publications (2015).
- Pavuluri, C.M., Kawamura, K., Swaminathan, T., Time-resolved distributions of bulk parameters, diacids, ketoacids and α -dicarbonyls and stable carbon and nitrogen isotope ratios of TC and TN in tropical Indian aerosols: Influence of land/sea breeze and secondary processes, *Atmospheric Research* **153**(2015), pp. 188-199.
- Pease, P.P., Tchakerian, V.P., Tindale, N.W., Aerosols over the Arabian Sea: geochemistry and source areas for aeolian desert dust, *Journal of Arid Environments* **39**(1998), pp. 477-496.
- Ponczek, M., Hayeck, N., Emmelin, C., George, C., Heterogeneous photochemistry of dicarboxylic acids on mineral dust, *Atmospheric Environment* **212**(2019), pp. 262-271.
- Rapf, R.J., Perkins, R.J., Carpenter, B.K., Vaida, V., Mechanistic Description of Photochemical Oligomer Formation from Aqueous Pyruvic Acid, *The Journal of Physical Chemistry A* **121**(2017), pp. 4272-4282.
- Rastogi, N., Sarin, M.M., Chemistry of aerosols over a semi-arid region: Evidence for acid neutralization by mineral dust, *Geophysical Research Letters* **33**(2006).
- Reed Harris, A.E. *et al.*, Photochemical Kinetics of Pyruvic Acid in Aqueous Solution, *The Journal of Physical Chemistry A* **118**(2014), pp. 8505-8516.
- Schaefer, T., van Pinxteren, D., Herrmann, H., Multiphase Chemistry of Glyoxal: Revised Kinetics of the Alkyl Radical Reaction with Molecular Oxygen and the Reaction of Glyoxal with OH, NO₃, and SO₄⁻ in Aqueous Solution, *Environmental Science & Technology* **49**(2015), pp. 343-350.
- Shen, M. *et al.*, Distribution and stable carbon isotopic composition of dicarboxylic acids, ketocarboxylic acids and α -dicarbonyls in fresh and aged biomass burning aerosols, *Atmos. Chem. Phys.* **22**(2022), pp. 7489-7504.
- Simoneit, B.R.T., Biomass burning-a review of organic tracers for smoke from incomplete combustion, *Appl. Geochem.* **17**(2002), pp. 129-162.
- Sorooshian, A. *et al.*, Oxalic acid in clear and cloudy atmospheres: Analysis of data from International Consortium for Atmospheric Research on Transport and Transformation 2004, *Journal of Geophysical Research: Atmospheres* **111**(2006).
- Stein, A. *et al.*, NOAA's HYSPLIT atmospheric transport and dispersion modeling system, *Bulletin of the American Meteorological Society* **96**(2015), pp. 2059-2077.

- Stephanou, E.G., Stratigakis, N., Oxocarboxylic and .alpha.,.omega.-dicarboxylic acids: photooxidation products of biogenic unsaturated fatty acids present in urban aerosols, *Environmental Science & Technology* **27**(1993), pp. 1403-1407.
- Tan, Y., Carlton, A.G., Seitzinger, S.P., Turpin, B.J., SOA from methylglyoxal in clouds and wet aerosols: Measurement and prediction of key products, *Atmospheric Environment* **44**(2010), pp. 5218-5226.
- Tan, Y., Perri, M.J., Seitzinger, S.P., Turpin, B.J., Effects of Precursor Concentration and Acidic Sulfate in Aqueous Glyoxal–OH Radical Oxidation and Implications for Secondary Organic Aerosol, *Environmental Science & Technology* **43**(2009), pp. 8105-8112.
- Tindale, N.W., Pease, P.P., Aerosols over the Arabian Sea: Atmospheric transport pathways and concentrations of dust and sea salt, *Deep Sea Research Part II: Topical Studies in Oceanography* **46**(1999), pp. 1577-1595.
- Wang, G. *et al.*, Field observation on secondary organic aerosols during Asian dust storm periods: Formation mechanism of oxalic acid and related compounds on dust surface, *Atmospheric Environment* **113**(2015), pp. 169-176.
- Wang, H., Kawamura, K., Yamazaki, K., Water-Soluble dicarboxylic acids, ketoacids and dicarbonyls in the atmospheric aerosols over the southern ocean and western pacific ocean, *Journal of Atmospheric Chemistry* **53**(2006), pp. 43-61.
- Xue, H., Khalizov, A.F., Wang, L., Zheng, J., Zhang, R., Effects of Coating of Dicarboxylic Acids on the Mass–Mobility Relationship of Soot Particles, *Environmental Science & Technology* **43**(2009), pp. 2787-2792.
- Yang, J. *et al.*, Molecular and spatial distributions of dicarboxylic acids, oxocarboxylic acids, and α -dicarbonyls in marine aerosols from the South China Sea to the eastern Indian Ocean, *Atmospheric Chemistry and Physics* **20**(2020a), pp. 6841-6860.
- Yang, J. *et al.*, Molecular and spatial distributions of dicarboxylic acids, oxocarboxylic acids, and α -dicarbonyls in marine aerosols from the South China Sea to the eastern Indian Ocean, *Atmos. Chem. Phys.* **20**(2020b), pp. 6841-6860.
- Yang, L., Ray, M.B., Yu, L.E., Photooxidation of dicarboxylic acids—Part II: Kinetics, intermediates and field observations, *Atmospheric Environment* **42**(2008), pp. 868-880.
- Yu, J.Z., Huang, X.-F., Xu, J., Hu, M., When Aerosol Sulfate Goes Up, So Does Oxalate: Implication for the Formation Mechanisms of Oxalate, *Environmental Science & Technology* **39**(2005), pp. 128-133.
- Zarzana, K.J. *et al.*, Primary emissions of glyoxal and methylglyoxal from laboratory measurements of open biomass burning, *Atmos. Chem. Phys.* **18**(2018), pp. 15451-15470.
- Zhang, Y. *et al.*, On-road vehicle emissions of glyoxal and methylglyoxal from tunnel tests in urban Guangzhou, China, *Atmospheric Environment* **127**(2016), pp. 55-60.

Table 1. Summarized statistical parameters describing the concentration levels of measured dicarboxylic acid, pyruvic acid, ω -oxocarboxylic acid in marine aerosols collected over the Arabian Sea during SS379 cruise (6-24 December 2018).

Compound name	Notation	Compound	Min-Max	Mean \pm Sd	Median
Oxalic acid	C ₂	HOOC-COOH	15.03-352.14	179.98 \pm 105.74	179.64
Malonic acid	C ₃	HOOC-CH ₂ -COOH	6.01-33.64	20.64 \pm 8.23	20.67
Succinic acid	C ₄	HOOC-(CH ₂) ₂ -COOH	8.13-62.38	32.32 \pm 12.85	32.61
Glutaric acid	C ₅	HOOC-(CH ₂) ₃ -COOH	1.55-22.16	7.94 \pm 4.65	7.77
Adipic acid	C ₆	HOOC-(CH ₂) ₄ -COOH	3.05-30.48	8.01 \pm 6.25	6.65
Pimelic acid	C ₇	HOOC-(CH ₂) ₅ -COOH	0.74-43.34	6.68 \pm 10.16	4.03
Suberic acid	C ₈	HOOC-(CH ₂) ₆ -COOH	0.16-0.91	0.41 \pm 0.2	0.4
Azelaic acid	C ₉	HOOC-(CH ₂) ₇ -COOH	5.36-63.71	19.63 \pm 14.94	14.64
Sebacic acid	C ₁₀	HOOC-(CH ₂) ₈ -COOH	0.28-29.87	2.98 \pm 7.05	0.87
Undecane dioic acid	C ₁₁	HOOC-(CH ₂) ₉ -COOH	2.04-21.32	8.58 \pm 4.69	7.5
Dodecanedioic acid	C ₁₂	HOOC-(CH ₂) ₁₀ -COOH	0.24-4.51	0.99 \pm 1.04	0.67
Methylmalonic acid	iC ₄	HOOC-CH(CH ₃)-COOH	0.18-32.59	2.84 \pm 7.7	0.69
Methylsuccinic acid	iC ₅	HOOC-CH ₂ (CH ₃)-COOH	1-10.03	4.08 \pm 2.3	3.58
Ketomalonic acid	kC ₃	HOOC-CHC(O)-COOH	1.48-5.51	2.9 \pm 1.84	2.31
Methylmaleic acid	mM	HOOC-(CH ₃)C=CH-COOH	0.31-7.46	1.58 \pm 1.62	1.29
Maleic acid (cis)	M	HOOC-HC=CH-COOH	0.44-2.68	1.41 \pm 0.65	1.19
Fumaric acid (trans)	F	HOOC-HC=CH-COOH	0.28-4.7	1.5 \pm 1.28	1.04
Ketopimelic acid	kC ₇	HOOC-(CH ₂) ₂ -C(O)-(CH ₂) ₂ -COOH	0.23-5.58	4.06 \pm 1.41	4.69
Phthalic acid	Ph	C ₆ H ₄ -(COOH) ₂	10.35-80.01	26.75 \pm 18.32	18.68
Isophthalic acid	iPh	C ₆ H ₄ -(COOH) ₂	0.63-5.71	2.41 \pm 1.28	2.13
Terephthalic acid	tPh	C ₆ H ₄ -(COOH) ₂	3.98-18.6	12.8 \pm 3.52	12.8
Pyruvic acid	Pyr	CH ₃ -C(O)-COOH	1.64-192.26	24.05 \pm 49.88	8.36
Glyoxylic acid	C ₂ ω	OHC-COOH	2.67-38.23	14.96 \pm 8.9	13.67
3-Oxopropanoic acid	C ₃ ω	OHC-(CH ₂)-COOH	0.33-15.8	3.88 \pm 3.58	3.06
4-Oxobutanoic acid	C ₄ ω	OHC-(CH ₂) ₂ -COOH	0.31-129.06	17.13 \pm 32.56	8.33
5-Oxopentanoic acid	C ₅ ω	OHC-(CH ₂) ₃ -COOH	0.12-2.7	0.96 \pm 0.72	0.75
7-Oxoheptanoic acid	C ₇ ω	OHC-(CH ₂) ₅ -COOH	2.62-10.58	6.19 \pm 2.04	5.63
8-Oxo-octanoic acid	C ₈ ω	OHC-(CH ₂) ₆ -COOH	1.22-12.24	5.83 \pm 4.2	4.77
Glyoxal	Gly	OHC-CHO	0.88-5.02	2.75 \pm 1.09	2.7
Methylglyoxal	MeGly	CH ₃ C(O)-CHO	0.07-3.18	1.31 \pm 1.06	1.15

Table 2. Regression slope between oxalic acid or oxalate (C_2) and $nss-SO_4^{2-}$ in the ambient aerosols, updated comparison after Yu et al. (2005).

Region	Probable Source	Sampling period	aerosol type	slope of oxalic acid (y) vs. $nss-SO_4^{2-}$ (x)	Reference
Arabian Sea	Indo-Gangetic Plain	6-24 Dec. 2018	TSP	0.018, $R^2 = 0.40$	This study
Bay of Bengal	Indo-Gangetic Plain	27 Dec. 2008-10 Jan. 2009	PM _{2.5}	0.0068, $R^2 = 0.64$	<u>Bikkina et al. (2017)</u>
Bay of Bengal	Southeast Asia	11-26 Jan. 2009	PM _{2.5}	0.006, $R^2 = 0.69$	<u>Bikkina et al. (2017)</u>
Hong Kong	East Asian pollution	Oct.-Nov. 2001	PM _{2.5}	0.034, $R^2 = 0.73$	<u>Yu et al. (2005)</u>
Pearl River Delta	East Asian pollution	Oct. 2002-Jun. 2003	PM _{2.5}	0.034, $R^2 = 0.73$	<u>Yu et al. (2005)</u>
Guangzhou, China	East Asian pollution	Jan. 2003	TSP	0.033, $R^2 = 0.88$	<u>Yu et al. (2005)</u>
South China Sea	East Asian pollution	Jan. 2003	TSP	0.050, $R^2 = 0.83$	<u>Yu et al. (2005)</u>
Nanjing, China	East Asian pollution	Feb'2001	PM _{2.5}	0.017, $R^2 = 0.70$	<u>Yu et al. (2005)</u>
Beijing, China	East Asian pollution	July-Oct'2002	PM _{2.5}	0.019, $R^2 = 0.91$	<u>Yu et al. (2005)</u>
Beijing, China	East Asian pollution	Jan'2003	PM _{2.5}	0.0046, $R^2 = 0.69$	<u>Yu et al. (2005)</u>
Jeju Island, Korea	East Asian pollution	April'2001	PM _{2.5}	0.047, $R^2 = 0.95$	<u>Yu et al. (2005)</u>
Nanjing, China	East Asian pollution	Sept'2001	PM _{2.5}	0.029, $R^2 = 0.70$	<u>Yu et al. (2005)</u>

Table 3. Percentage contributions of oxalic acid, total dicarboxylic acids, oxocarboxylic acids (including pyruvic acid), and α -dicarbonyls to water-soluble organic carbon (WSOC), organic carbon (OC), and total carbon (TC) levels in marine aerosols collected over the Arabian Sea during SS379 cruise (6-24 December 2018).

Compound class	Min-Max	Mean \pm Sd	Median
Oxalic acid-C/WSOC (%)	0.2-5.6	1.6 \pm 1.3	1.3
Oxalic acid-C/OC (%)	0.1-2.1	0.8 \pm 0.5	0.8
Oxalic acid-C/TC (%)	0.1-1.7	0.6 \pm 0.4	0.7
Dicarboxylic acids/WSOC (%)	1.2-21	4.6 \pm 4.6	3.2
Oxocarboxylic acids/WSOC (%)	0.3-4.5	1.1 \pm 1.3	0.6
α -Dicarbonyls/WSOC (%)	0.02-0.47	0.07 \pm 0.11	0.04
Dicarboxylic acids/OC (%)	0.7-4.4	2 \pm 0.9	1.6
Oxocarboxylic acids/OC (%)	0.1-2.6	0.5 \pm 0.6	0.2
α -Dicarbonyls/OC (%)	0.01-0.07	0.03 \pm 0.01	0.02
Dicarboxylic acids/TC (%)	0.7-3.5	1.6 \pm 0.7	1.6
Oxocarboxylic acids/TC (%)	0.1-2.1	0.4 \pm 0.5	0.2
α -dicarbonyls/TC (%)	0.01-0.06	0.02 \pm 0.01	0.02

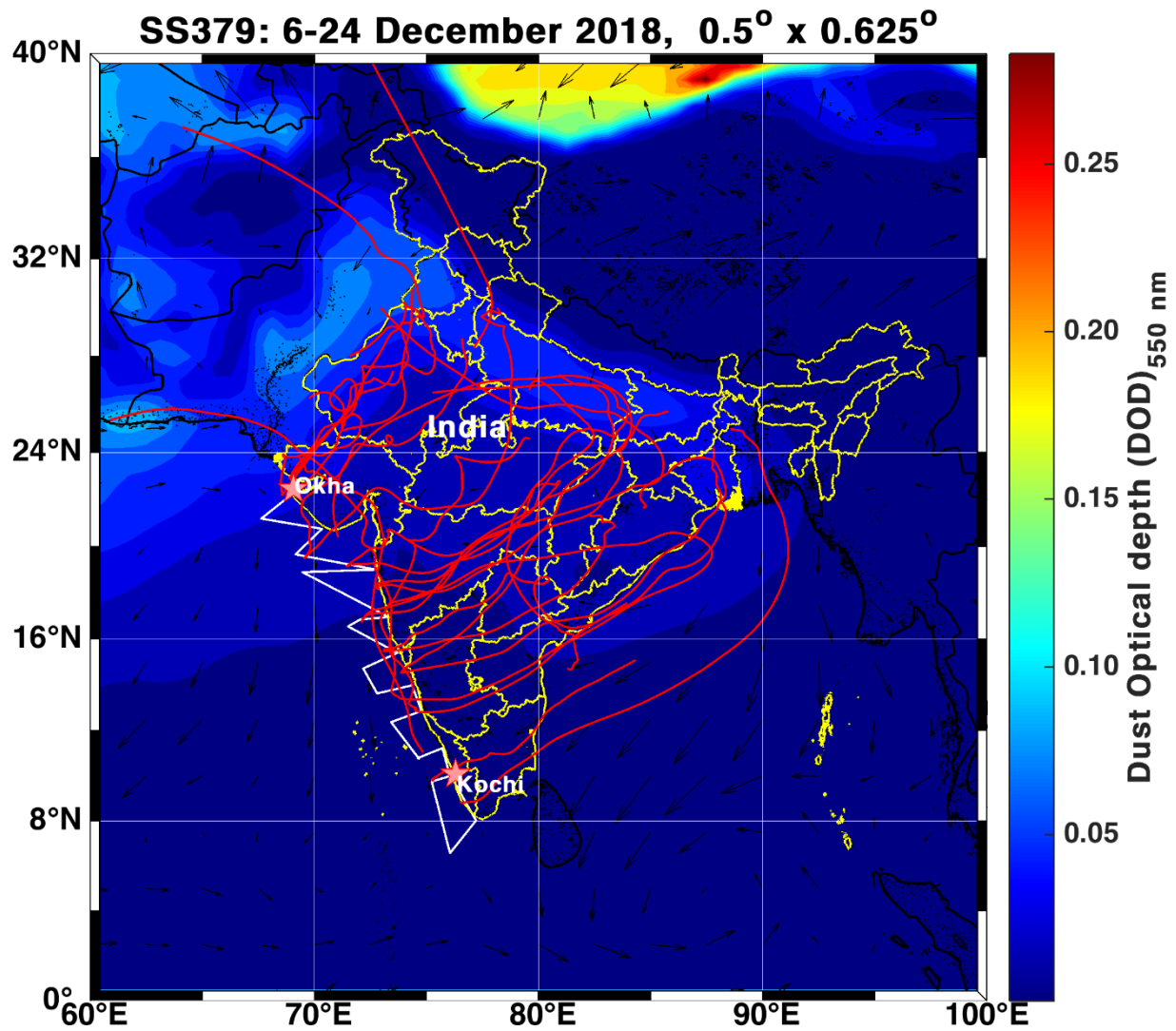


Fig. 1. Dust aerosol optical depth retrievals at 550 nm from the MODIS satellite during SS379 cruise in the Arabian Sea. 7-day air mass back trajectories computed from the HYSPLIT model for the TSP sampling days over the Arabian Sea

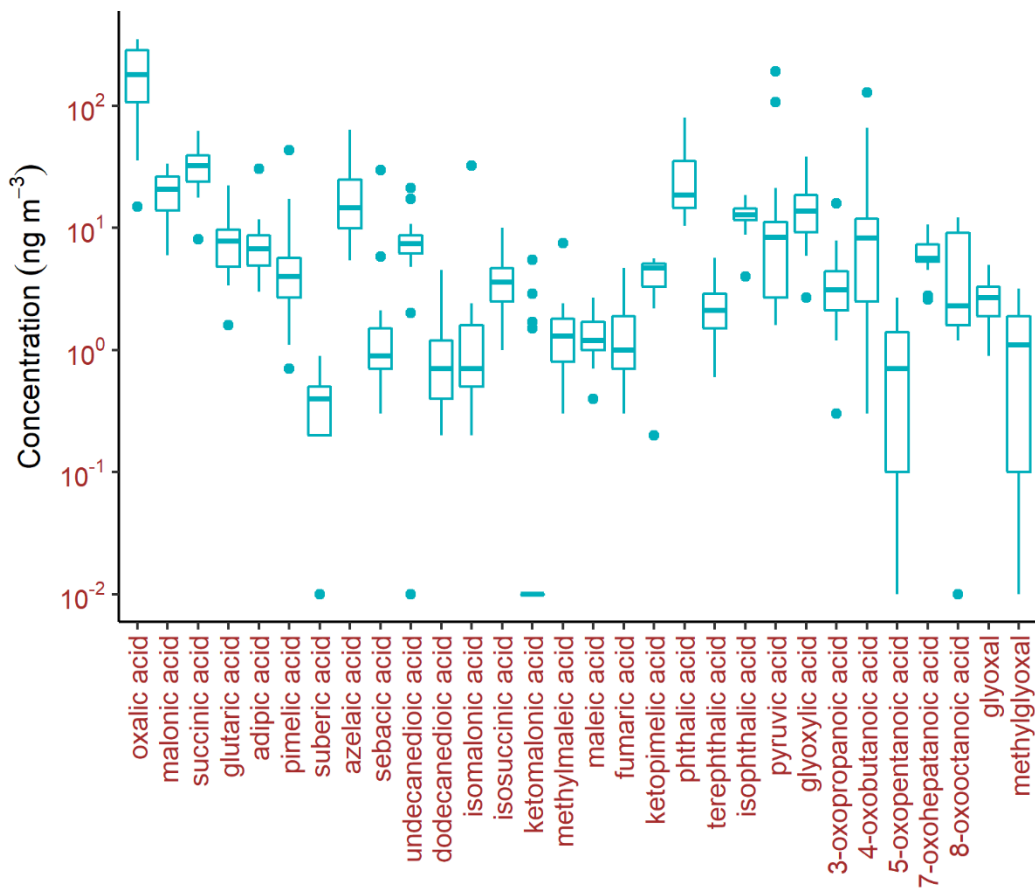


Fig. 2. Box-whisker plots of concentrations of measured aliphatic, multifunctional, unsaturated aliphatic and aromatic dicarboxylic acids, pyruvic acid, ω -oxocarboxylic acids, and α -dicarbonyls in marine aerosols collected over the Arabian Sea during SS379 cruise.

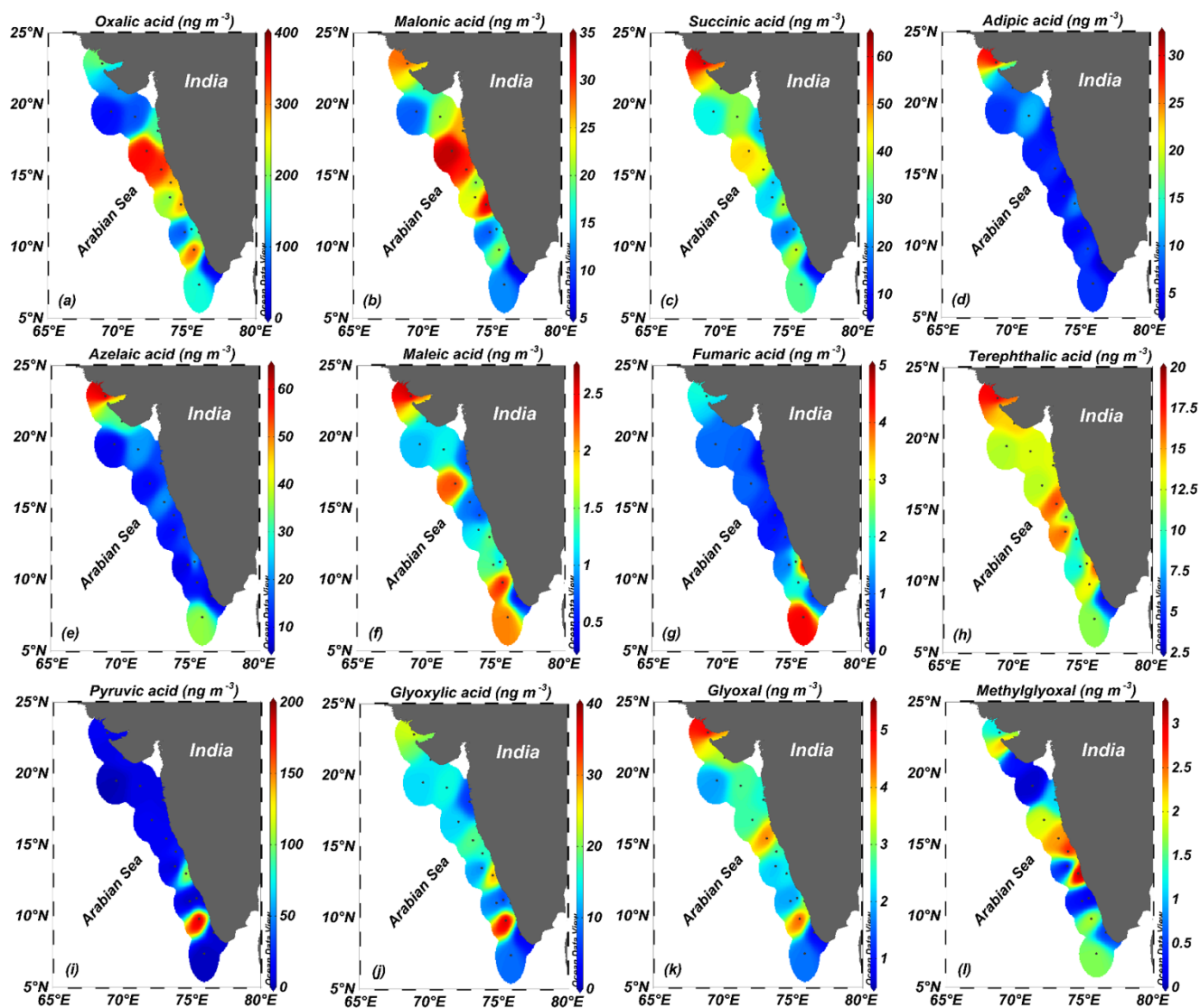


Fig. 3. Spatial variability of concentrations of important (a-h) dicarboxylic acids, (i) pyruvic acid, (j) glyoxylic acid and (k-l) α -dicarbonyls in marine aerosols collected over the Arabian Sea during SS79 cruise (6-24 December 2018).

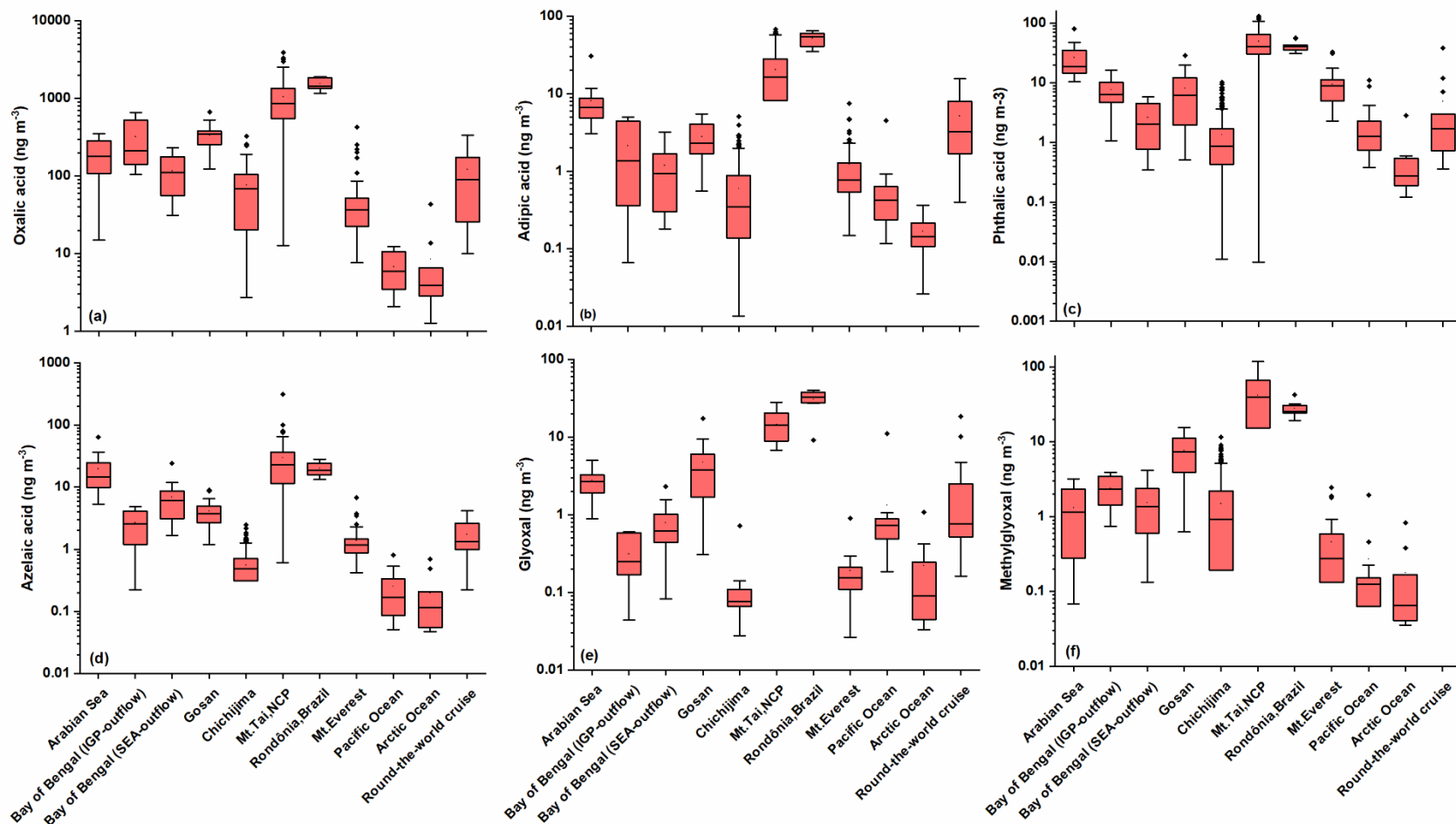


Fig. 4. A comparison of concentrations of (a) oxalic acid, (b) adipic acid, (c) phthalic acid, (d) azelaic acid, (e) glyoxal, (f) methylglyoxal in marine aerosols collected over the Arabian Sea during the impact of continental outflow with those from the Bay of Bengal, Gosan, Chichijima, Rondônia (Brazil), Mt. Everest, Pacific Ocean, Arctic Ocean and round-the-world cruise (Fu et al., 2013; Cong et al., 2015; Bikkina et al., 2017).

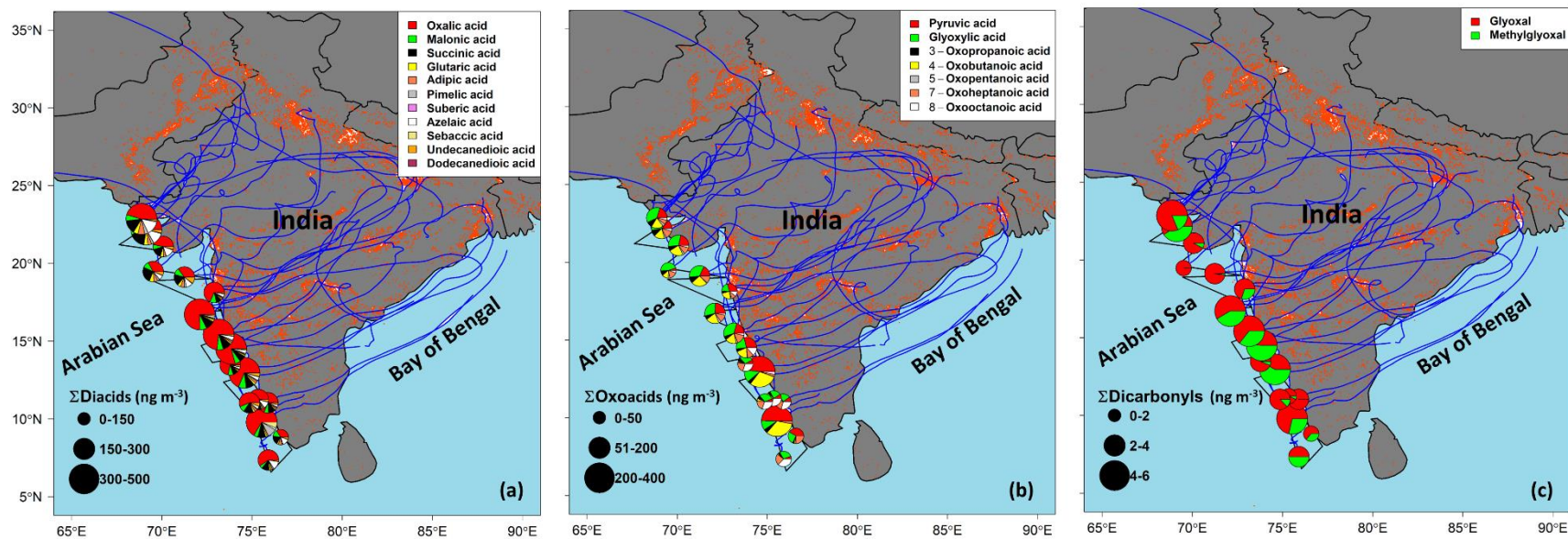


Fig. 5. Spatial variability of molecular distributions of (a) aliphatic dicarboxylic acids, (b) oxocarboxylic acids including pyruvic acid, and (c) α -dicarbonyls in marine aerosols collected over the Arabian Sea during the SS379 cruise (6-24 December 2018).

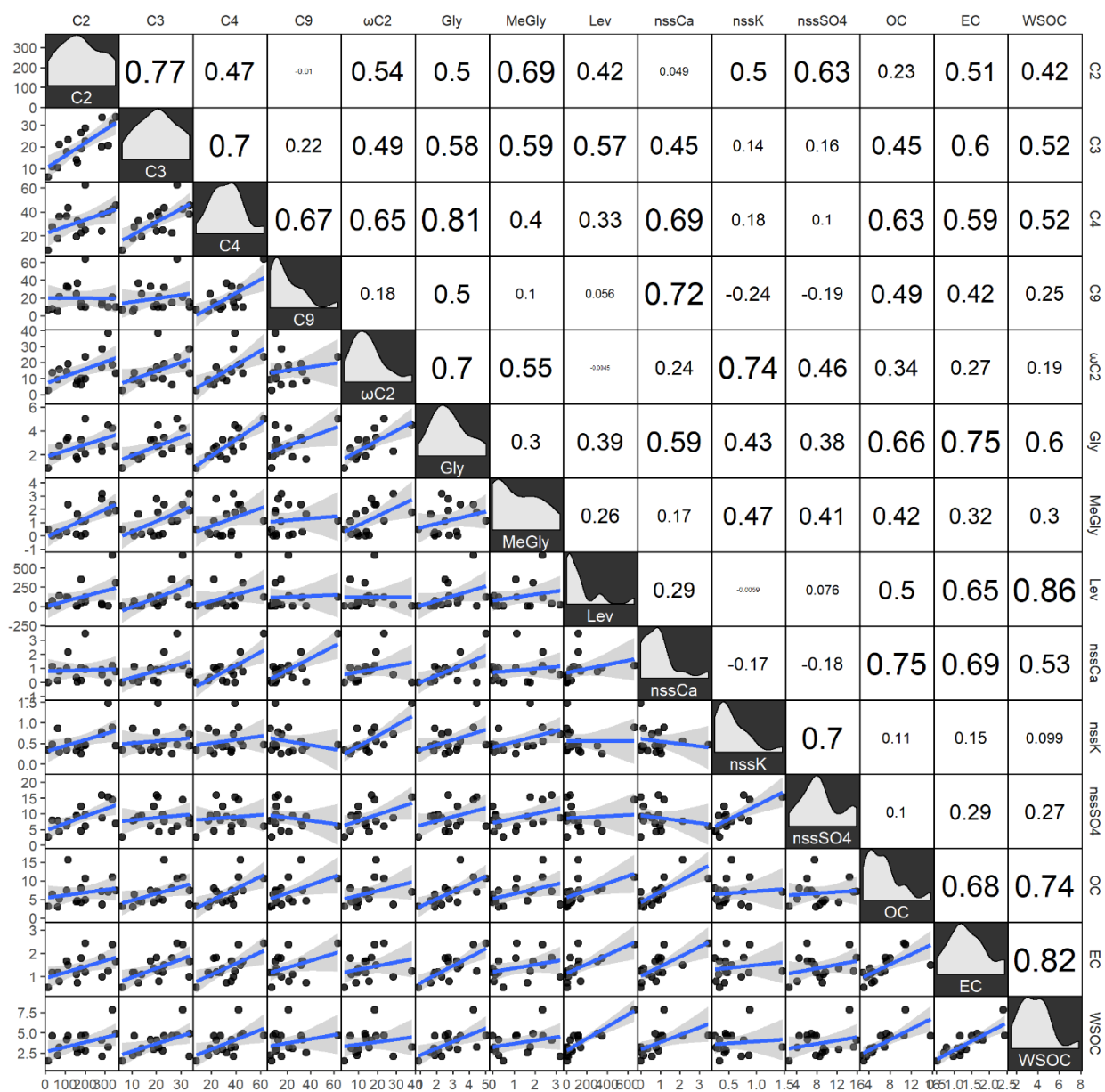


Fig. 6. Correlation matrix of LMW-dicarboxylic acids (oxalic acid: C₂, malonic acid: C₃, succinic acid: C₄) and azelaic acid (C₉) with other major continental tracers including glyoxylic acid (ω C₂), glyoxal (Gly) and methylglyoxal (MeGly), levoglucosan (*Lev* nss-Ca²⁺, nss-K⁺, nss-SO₄²⁻, OC, EC, and WSOC in marine aerosols collected over the Arabian Sea during SS379 cruise (6-24 December 2018). The diagonal elements correspond to probability distribution curves for the parameter of choice and the lower and upper diagonal elements represent the linear regression analysis and corresponding Pearson's correlation coefficient values.

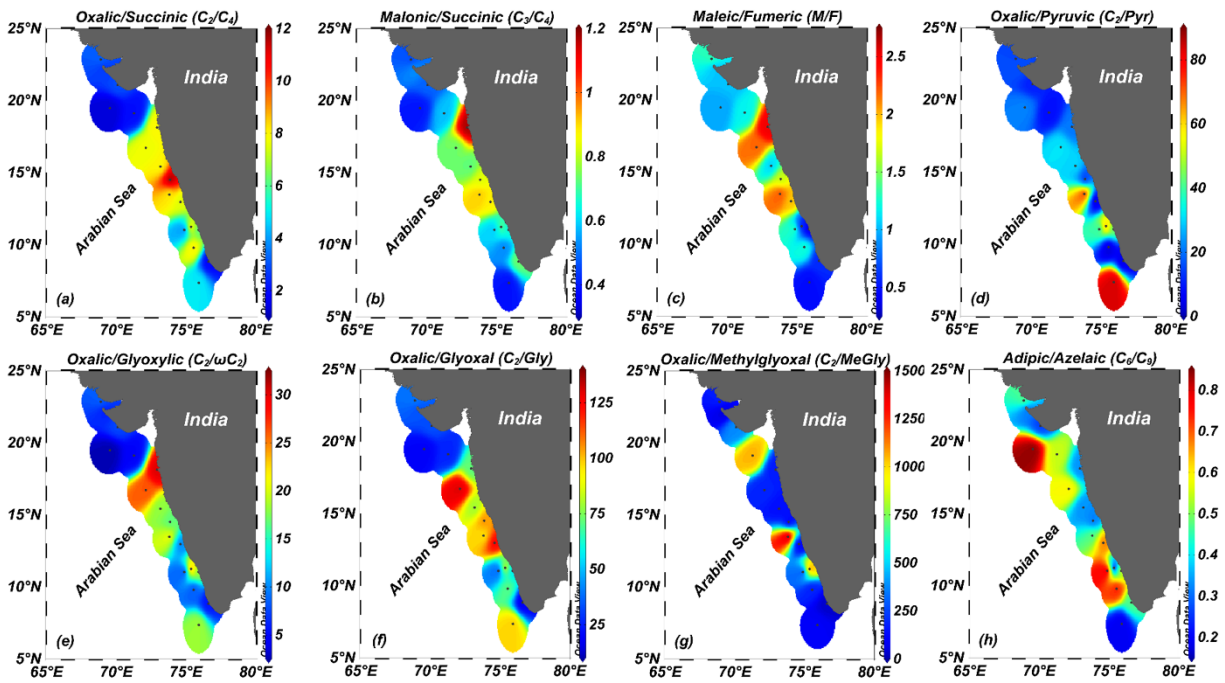


Fig. 7. Spatial variability of diagnostic concentration ratios of dicarboxylic acids and related polar compounds over the Arabian Sea during SS379 cruise (6-24 December 2018)

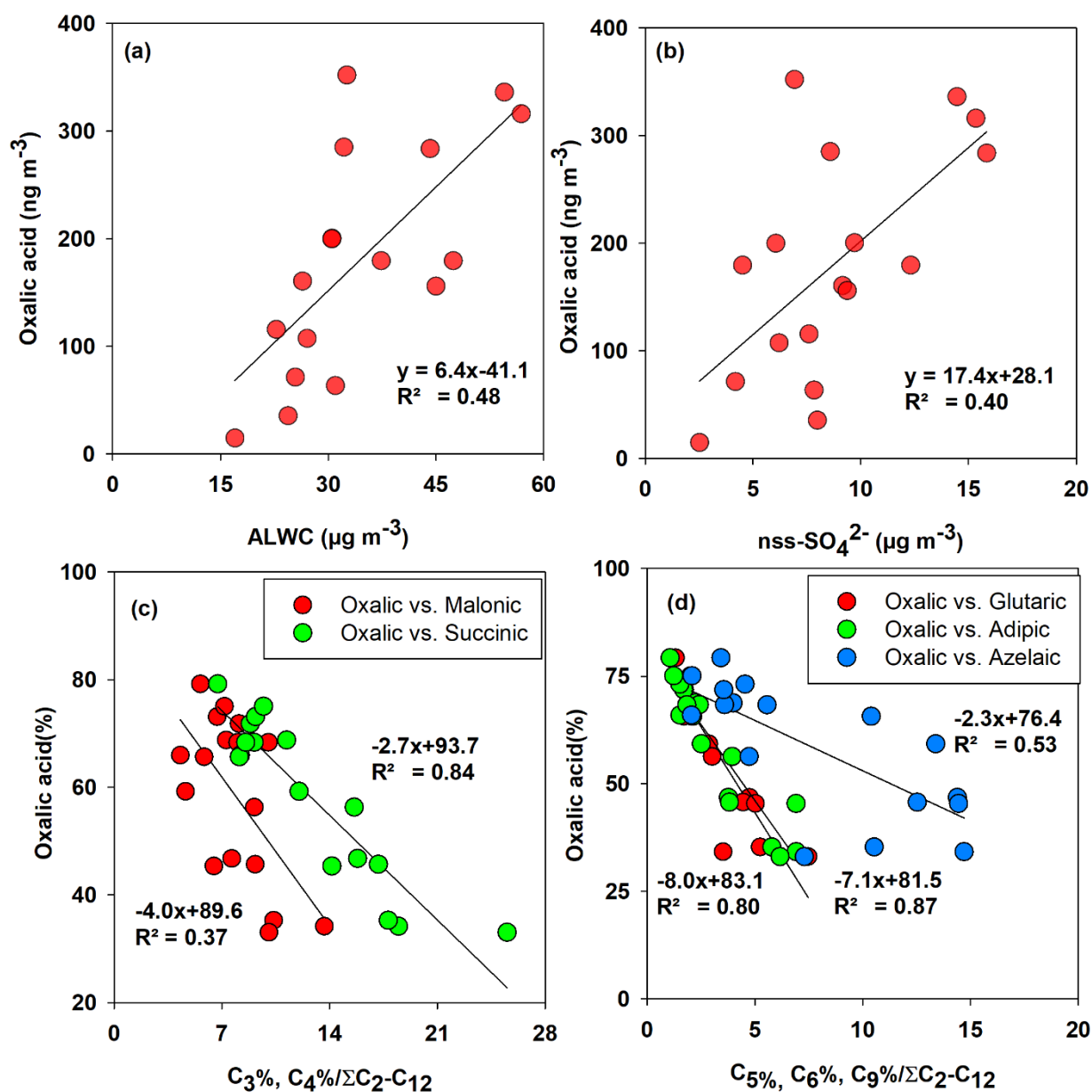


Fig. 8. Linear regression analysis between the concentrations of oxalic acid (C_2) and (a) aerosol liquid water content (ALWC), and, (b) $nss-SO_4^{2-}$ over the Arabian Sea during SS37 cruise (6-24 December 2018). Panels (c) and (d) refer to relative abundance of C_2 in WSOC versus % abundance ratio of C_3 , C_4 , C_5 , C_6 , C_9 in their total aliphatic dicarboxylic acids in marine aerosols collected over the Arabian Sea during SS379 cruise

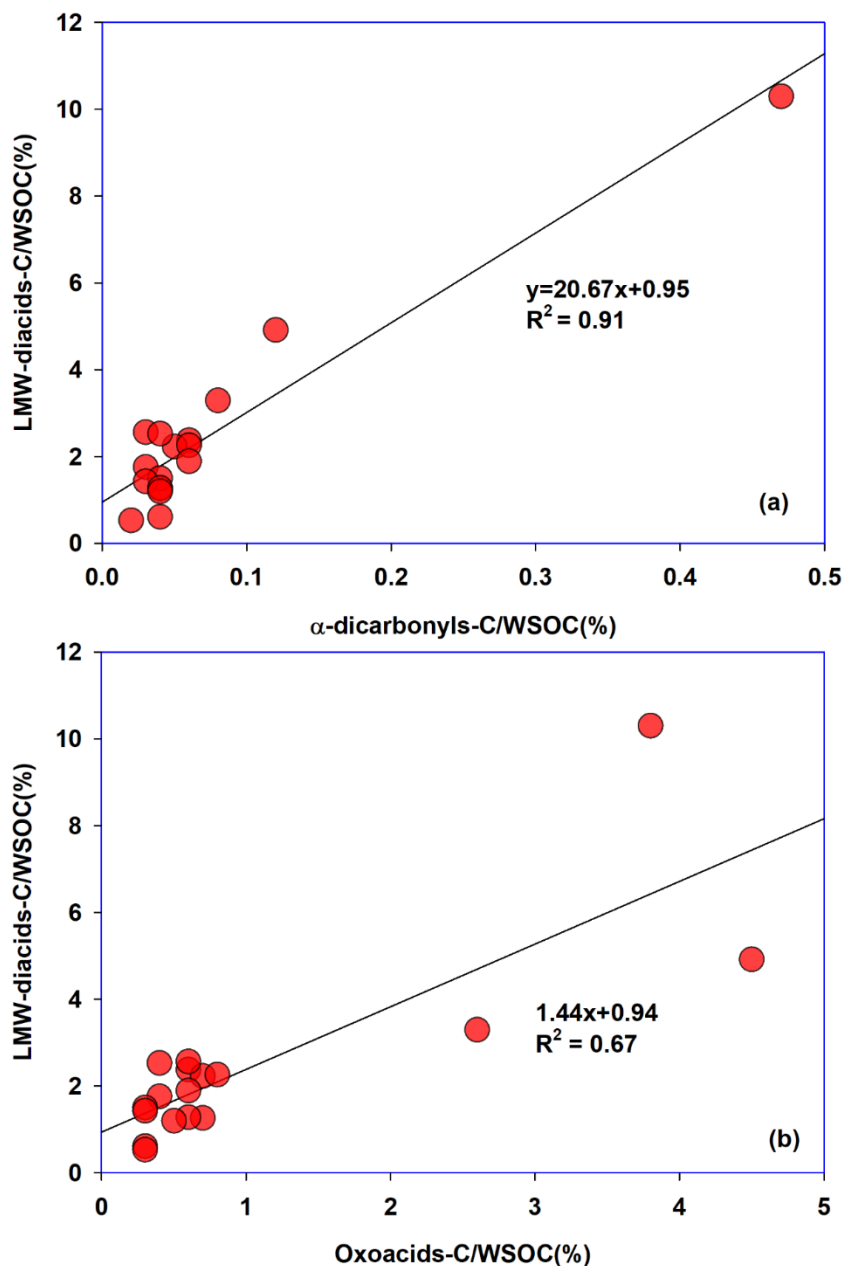


Fig. 9. Linear regression analysis between the percentage relative abundances of (a) α -dicarbonyls and (b) oxoacids in WSOC versus that of LMW-diacids in WSOC in marine aerosols collected over the Arabian Sea during SS379 cruise (6-24 December 2022).

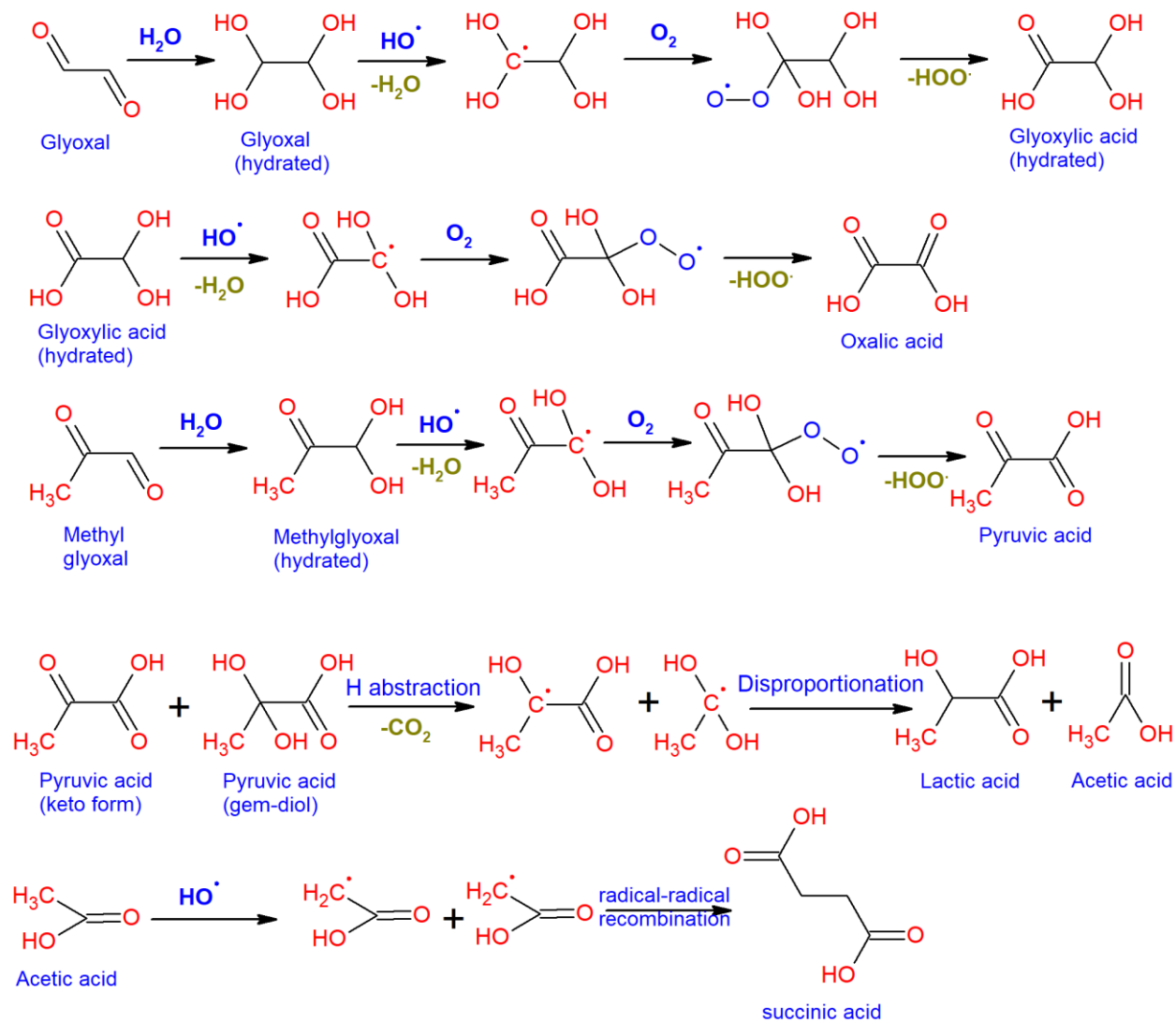


Fig. 10. Schematic representation for formation pathways of oxalic acid and related water-soluble compounds in marine aerosols collected over the Arabian Sea during the impact of South Asian outflow.