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Supplement of

**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**

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Linear correlations

20 In order to investigate the origins and formation pathways of diacids and related compounds in marine aerosols, we examined the correlations between selected species and also conducted linear regression analyses for mass ratios of the selected chemical species. Moderate correlations were observed for C₆ with C₉ diacids in the EIO-WI and EIO-SL, and significant correlations over the SCS and Malacca regions. Such positive correlations indicate that the C₆ diacid should have also been significantly generated from the biogenic VOCs such as cyclic olefins, in
25 addition to the anthropogenic sources.

Hydroxy succinic acid (malic acid, hC₄), C₄ and C₃ diacids in the SCS correlated well with each other ($p < 0.05$; $R^2 = 0.87$ for hC₄ vs C₃ and $R^2 = 0.78$ for hC₄ vs C₄; Fig. S2a and S2b). The formation of C₃ diacid from C₄ diacid through hC₄ as an intermediate has been proposed by Kawamura and Sakaguchi (1999) and also observed in a laboratory study (Yang et al., 2008). A very good linear relationship of hC₄ diacid with C₄ diacid ($R^2 = 0.85$) and
30 C₂ diacid ($R^2 = 0.78$) has been reported in forest aerosols from central Europe, which have been attributed to *in-situ* photochemical production from biogenic unsaturated fatty acids (Kourtchev et al., 2009). Therefore, the good correlations observed among C₃, C₄ and hC₄ in the SCS implies that they were derived from common but mixed (both biogenic and anthropogenic) sources and the *in-situ* secondary formation might be more intensive, rather than the transformations from C₄ to C₃ via hC₄ diacids. A significant linear correlation was also found between
35 methylmalonic (iC₄) acid with C₃ and C₄ diacids. The branched chain diacids could come from the oxidation of isoprene released from the ocean surface. Such linear correlations also suggest that the diacids and related compounds were photochemical oxidation products of isoprene and biogenic unsaturated fatty acids in the MABL, and also confirm the intensive *in-situ* secondary formation of diacids and related species. A notable feature observed here is the significant ($p < 0.05$) correlations between MeGly, Pyr, and ω C₂ with C₃ and C₄ diacids over Malacca, but not in other regions. Nguyen et al. (2010) found that C₃ and C₄ diacids were formed from isoprene oxidized by
40 ozone based on the laboratory experiment. Furthermore, it has been reported that isoprene emission results in significant enhancement of SOA and O₃ levels at coastal U.S. sites (Gantt et al., 2010). Hence, the loading of C₃ and C₄ diacids also should have been influenced by not only the unsaturated fatty acids but also the isoprene, at least in Malacca region.

45 The C₂/(Σ C₂-C₁₀) ratios showed negative relationships with C₃ and C₄ diacids over the SCS to EIO (Fig. S3a and S3b). This phenomenon again confirms the formation of C₂ diacid from C₃ and C₄ diacids by their photochemical transformations during long-range atmospheric transport. It is also worth noting that the C₄/(Σ C₂-C₁₀) ratios showed a linear relationship with glutaric (C₅) acid in all regions, except the EIO-WI (Fig. S3c). Additionally, a similar variation trend is observed for the concentrations of C₅ and C₉ diacids in all regions, although

50 they were not significant (C_9 ; Fig. S3d). Such positive relations reveal that they all should have been simultaneously
produced significantly by in-situ photochemical reactions over the oceanic region, although we do not preclude the
photochemical transformations of long-chain diacids to their lower homologues including C_6 - C_4 diacids. Thus, the
observed linear correlations among the relative abundances of C_2 , C_3 , C_4 , C_5 , and C_9 infer that the long-chain as
well as C_3 and C_4 diacids were mainly derived from biogenic emissions followed by the in-situ secondary reactions
55 whereas the C_2 diacid is mainly produced by the photochemical transformation of their higher over the SCS to EIO.
The long-range transported air masses from the continental region that should be enriched with the anthropogenic
emissions including oxidant species should have been promoted the oxidation of unsaturated fatty acids and
BVOCs, resulting the enhanced levels of diacids and related compounds over the remote oceanic region.

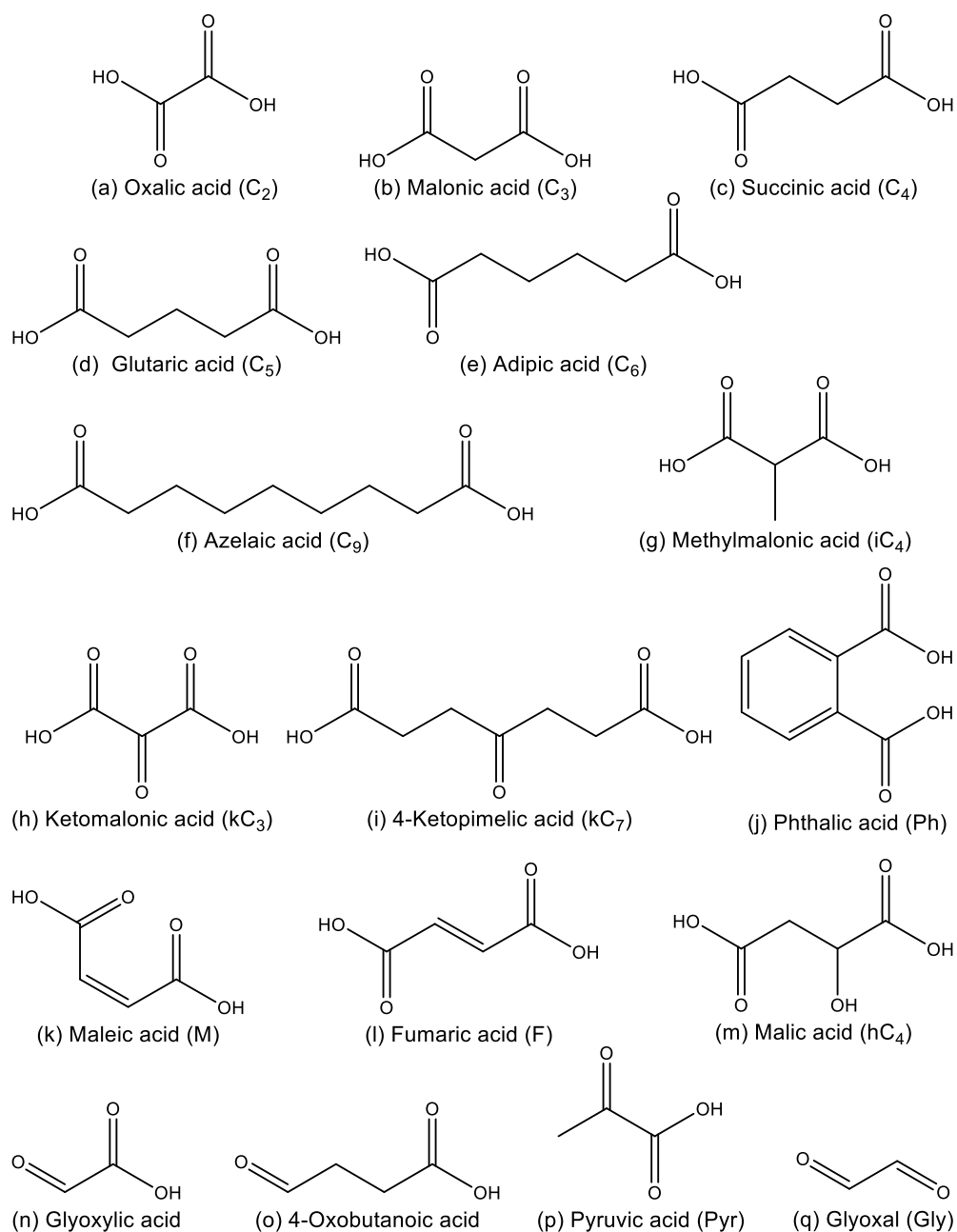
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Table S1. Correlation matrices of selected diacids and related compounds in TSP collected during the NORC2015-10 cruise.

Correlation	C ₂	C ₃	C ₄	C ₅	C ₆	C ₉	Ph	Pyr	ωC ₂	MeGly
SCS (n=27)										
C ₂	1.00									
C ₃	0.85 **	1.00								
C ₄	0.79 **	0.88 **	1.00							
C ₅	0.86 **	0.73 **	0.87 **	1.00						
C ₆	0.87 **	0.67 **	0.75 **	0.95 **	1.00					
C ₉	0.67 **	0.38	0.36	0.70 **	0.78 **	1.00				
Ph	0.70 **	0.54 **	0.72 **	0.86 **	0.84 **	0.59 **	1.00			
Pyr	0.50 **	0.44 *	0.55 **	0.52 **	0.43 *	0.19	0.52 **	1.00		
ωC ₂	0.88 **	0.71 **	0.68 **	0.80 **	0.79 **	0.68 **	0.69 **	0.66 **	1.00	
MeGly	0.65 **	0.40 *	0.30	0.42*	0.49 *	0.50 **	0.50 **	0.59 **	0.80 **	1.00
EIO-WI (n=20)										
C ₂	1.00									
C ₃	0.90 **	1.00								
C ₄	0.89 **	0.94 **	1.00							
C ₅	0.85 **	0.83 **	0.76 **	1.00						
C ₆	0.26	0.37	0.41	0.41	1.00					
C ₉	0.34	0.36	0.36	0.39	0.51 *	1.00				
Ph	0.11	0.29	0.18	0.51 *	0.67 **	0.40	1.00			
Pyr	0.10	0.13	0.14	0.17	0.04	0.33	0.04	1.00		
ωC ₂	0.68 **	0.51 *	0.53 *	0.71 **	-0.04	0.40	0.12	0.23	1.00	
MeGly	0.11	0.13	0.13	0.00	0.21	0.28	-0.11	0.61 **	-0.12	1.00
EIO-SL (n=28)										
C ₂	1.00									
C ₃	0.89 **	1.00								
C ₄	0.86 **	0.94 **	1.00							
C ₅	0.73 **	0.75 **	0.78 **	1.00						
C ₆	0.41 *	0.31	0.49 **	0.44	1.00					
C ₉	0.31	0.14	0.20	0.31	0.39 *	1.00				
Ph	0.53 **	0.35	0.43 *	0.52 **	0.70 **	0.79 **	1.00			
Pyr	0.40 *	0.20	0.38 *	0.24	0.38 *	0.22	0.27	1.00		
ωC ₂	0.79 **	0.55 **	0.65 **	0.61 **	0.50 **	0.51 **	0.69 **	0.64 **	1.00	
MeGly	0.21	0.21	0.09	0.11	0.14	0.07	0.19	0.32	0.14	1.00
Malacca (n=9)										
C ₂	1.00									
C ₃	0.65	1.00								
C ₄	0.45	0.88 **	1.00							
C ₅	0.69 *	0.76 *	0.87 **	1.00						
C ₆	0.89 **	0.64	0.56	0.85 **	1.00					
C ₉	0.87 **	0.71 *	0.60	0.81 **	0.96 **	1.00				
Ph	0.84 **	0.87 **	0.80 *	0.89 **	0.90 **	0.94 **	1.00			
Pyr	0.55	0.71 *	0.83 **	0.79 *	0.60	0.56	0.71 *	1.00		
ωC ₂	0.80 **	0.75 *	0.80 **	0.97 **	0.91 **	0.86 **	0.91 **	0.84 **	1.00	
MeGly	0.36	0.79 *	0.94 **	0.83 **	0.50	0.52	0.74 *	0.67	0.71 *	1.00

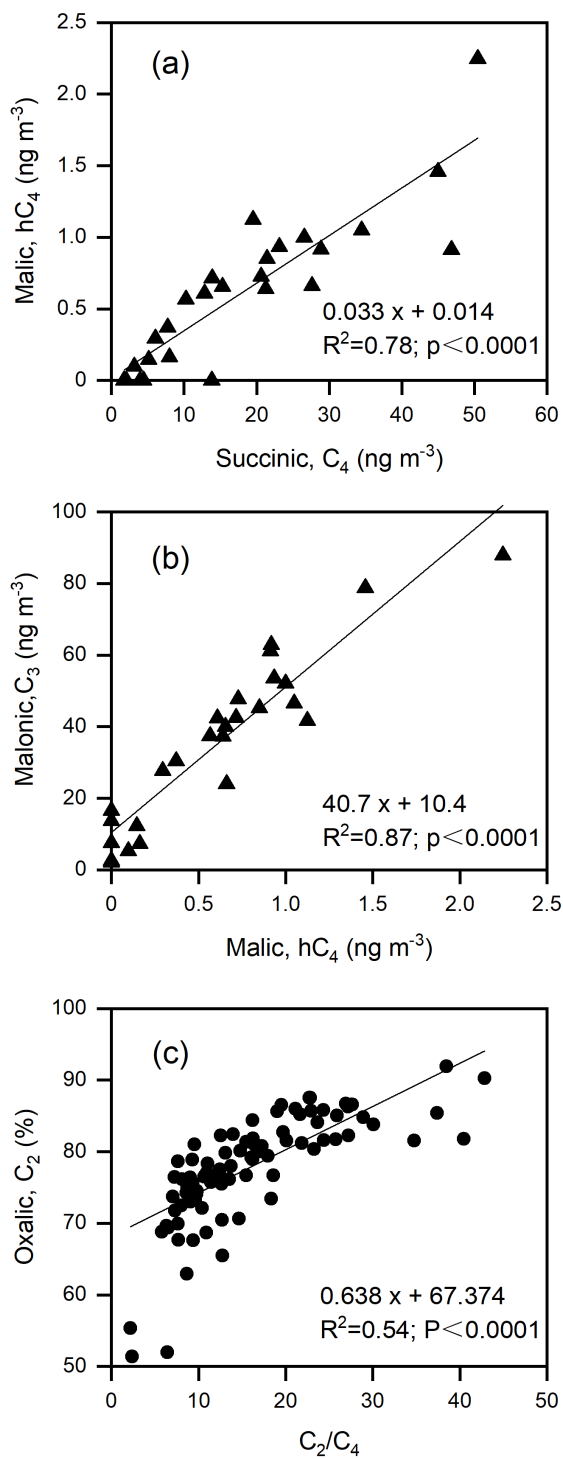
** : At 0.01 level (two-tailed), the correlation is significant.

* : At 0.05 level (two-tailed), the correlation is significant.



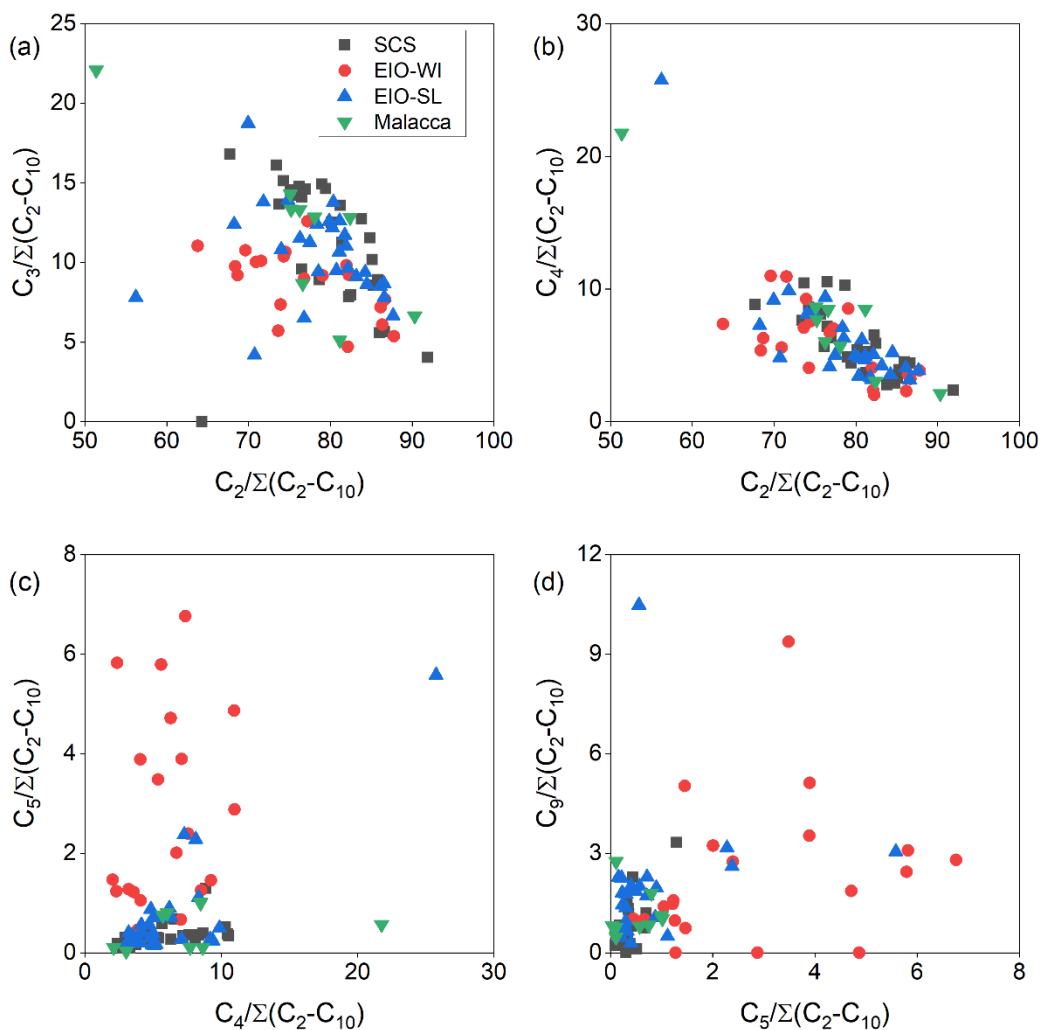
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Figure S1. Chemical structures of selected dicarboxylic acids and other major organic compounds detected in the marine aerosols.



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Figure S2. Scatterplots for (a) malic versus succinic acids, (b) malonic versus malic acids in aerosols collected over the SCS; (c) relative abundance of oxalic acid in total diacid mass versus C₂/C₄ ratios in aerosols collected over the all sea areas.



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Figure S3. Scatterplots between the relative abundances of C₂ in their total aliphatic homologues (ΣC_2-C_{10}) with that of (a) malonic acid and (b) succinic acid. Likewise, scatterplots showing the relative abundances of C₅ in ΣC_2-C_{10} with those of (c) C₄ and (d) C₉ diacids in TSP samples collected during the NORC2015-10 cruise.

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