

Response to Editor

Editor Decision: Publish subject to minor revisions (review by editor) (07 Feb 2020) by [Willy Maenhaut](#)

Comments to the Author:

The authors have reasonably well addressed the comments of the two anonymous referees and they have modified their manuscript accordingly. However, the comments given below should be addressed and several alterations are needed for the Main text and Supplement before the manuscript can be published in ACP.

Response: We thank the comments and revise our manuscript accordingly.

For the Main text:

Line 19: Replace "the snapshot" by "our snapshot".

Response: Done.

Line 20: Replace "to the mean" by "to a mean".

Response: Done.

Line 21: Replace "closer to" by "close to".

Response: Done.

Line 29: Replace "discussed the" by "discuss the".

Response: Done.

Line 51-54: Although SOA from the photo-oxidation of isoprene produced by phytoplankton blooms has been proposed as a potentially important marine SOA source by Meskhidze and Nenes (2006), a study by Claeys et al. (Chemical characterisation of marine aerosol at Amsterdam Island during the austral summer of 2006-2007, *J. Aerosol Sci.* 41 (2010) 13-22) found no evidence for isoprene SOA at that remote site. On the other hand, Gantt, Meskhidze, and Kamykowski (A new physically-based quantification of marine isoprene and primary organic aerosol emissions, *Atmos. Chem. Phys.* 9 (2009) 4915-4927) state "Using a fixed 3% mass yield for the conversion of isoprene to SOA, our emission simulations show minor (<0.2%) contribution of marine isoprene to the total marine source of OC on a global scale. However, our model calculations also indicate that over the tropical oceanic regions (30°S to 30°N), marine isoprene SOA may contribute over 30% of the total monthly-averaged sub-micron OC fraction of marine aerosol. The estimated contribution of marine isoprene SOA to hourly-averaged sub-micron marine OC emission is even higher, approaching 50% over the vast regions of the oceans during the midday hours when isoprene emissions are

34 highest". This is in contrast with the study by Arnold et al. (Evaluation of the global oceanic
35 isoprene source and its impacts on marine organic carbon aerosol, *Atmos. Chem. Phys.* 9
36 (2009) 1253-1262), where it is stated "Inclusion of secondary organic aerosol (SOA)
37 production from oceanic isoprene in the model with a 2% yield produces small contributions
38 (0.01-1.4%) to observed organic carbon (OC) aerosol mass at three remote marine sites in the
39 Northern and Southern Hemispheres. Based on these findings we suggest an insignificant role
40 for isoprene in modulating remote marine aerosol abundances, giving further support to a
41 recently postulated primary OC source in the remote marine atmosphere". Note also that the
42 study of Claeys et al. (2004) does not deal with marine isoprene SOA, but instead with
43 isoprene SOA from the Amazon Basin. Using "marine AND SOA AND isoprene" as topic on
44 the Web of Science I receive 47 hits. Therefore, the sentence here should be rewritten and
45 appropriate references should be given. It is in any case clear that isoprene emissions from
46 the continents are much more important than those from the oceans, as appears from the
47 paper by Guenther et al. (A global model of natural volatile organic compound emissions, *J.*
48 *Geophys. Res.* 100 (1995) 8873-8892), so that the importance of marine isoprene SOA should
49 be downplayed.

50 **Response:** Thanks. We revise this sentence into "Secondary organic aerosols (SOAs) arising
51 from the oxidation of phytoplankton-derived isoprene have been argued to affect the
52 chemical composition of marine atmospheric aerosols and consequently impact CCN loading
53 and cloud droplet number concentrations (Ekström et al., 2009; Meskhidze and Nenes, 2006),
54 but the importance of the marine isoprene-derived SOA is still debated (Arnold et al., 2009;
55 Claeys et al., 2010; Gantt et al., 2009; Guenther et al., 1995). For example, Gantt et al. (2009)
56 estimated the contribution of marine isoprene-derived SOA to the OC in marine atmospheric
57 particles ranged from <0.2% on a global scale, but to as high as 50% (sub-micron OC) over
58 the vast regions of the oceans during the midday hours when isoprene emissions are
59 highest." .

60 For the reference "(Claeys et al., 2004)", it was revised in "However, emission fluxes and
61 oxidation processes of BVOCs show great variation, depending on global warming and other
62 factors such as regional landscape, other pollutants in the ambient air, etc. (Ait-Helal et al.,
63 2014; Claeys et al., 2004; Hu and Yu, 2013; Peñuelas and Staudt, 2010)." in line 69-71.

64 Line 56: Replace "future warming climate in the future" by "future warming climate".

65 **Response:** Done.

66 Line 61: See what I wrote above about marine isoprene SOA; its importance should be
67 downplayed.

68 **Response:** Thanks. We revise this sentence into “More importantly, BVOCs emitted from
69 continental ecosystems and their oxidation products can significantly affect the atmosphere in
70 remote marine areas through long-range transport (Ding et al., 2013; Fu et al., 2011; Hu et al.,
71 2013a; Kang et al., 2018; Kawamura et al., 2017).”.

72 Line 66: Replace "emissions fluxes" by "emission fluxes".

73 **Response:** Done.

74 Line 68: Replace "air etc." by "air, etc.".

75 **Response:** Done.

76 Line 71: Replace "Sharma," by "Sharma et al.,".

77 **Response:** Done.

78 Line 74: I do not understand the use of "revere" here; should it perhaps be "reverse" instead
79 of "revere"?

80 **Response:** Done. We revise it into “reverse”.

81 Line 75: Replace "Update observations" by "Updated observations".

82 **Response:** Done.

83 Line 77: Replace "we analyzed the" by "we determined the".

84 **Response:** Done.

85 Line 119: Replace "run every" by "ran every".

86 **Response:** Done.

87 Line 132: Replace "under the controlled" by "under controlled".

88 **Response:** Done.

89 Line 142: Replace "the smaller difference among the means" by "the small difference among
90 the mean".

91 **Response:** Done.

92 Line 156: Replace "suggested that" by "suggest that".

93 **Response:** Done.

94 Line 170: Replace "indicated a" by "indicate a".

95 **Response:** Done.

96 Line 175: Replace "traceries and" by "trajectories and".

97 **Response:** Done.

98 Line 197: Replace "differences were found" by "difference was found".

99 **Response:** Done.

100 Line 198: Replace "limited samples" by "limited number of samples".

101 **Response:** Done.

102 Line 199: Replace "emissions sources" by "emission sources".

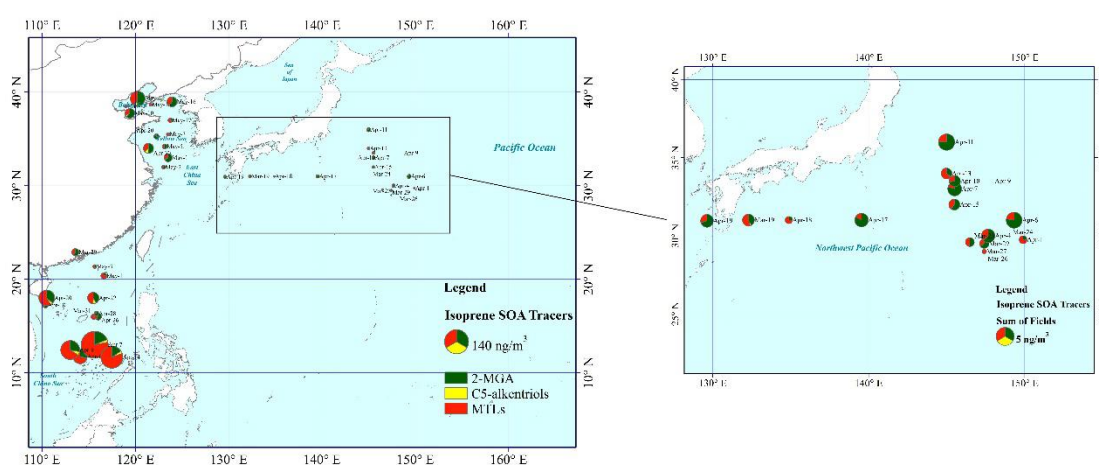
103 **Response:** Done.

104 Lines 230 and 233: I presume that it should be "SOAI" instead of "SOA".

105 **Response:** Done.

106 Page 9, Figure 3, right panel: Replace "SOA Traces" by "SOA Tracers".

107 **Response:** Done. And the revised figure is shown below.



108 Lines

109 240, 260, 408, 418, 420 and 436: It is unclear what "ECS" denotes; I presume that it stands

110 for "East China Sea". In any case, abbreviations and acronyms should be defined (written

111 full-out) when first used. Also, I think that "ECS" in two of these cases (i.e., in lines 240 and

112 436) should be replaced by "YBS".

113 **Response:** Thanks.

114 The "ECS" in line 240 and 436 was revised as "YBS". And abbreviations for "ECS" is

115 defined (written full-out, "East China Sea") when first used in line 263-264.

116 Line 248: Replace "atmospheres (Gordon" by "atmosphere (Gordon".

117 **Response:** Done.

118 Line 266: Replace "confirm" by "confirm this".

119 **Response:** Done.

120 Line 276: Replace "remains unexplained" by "remain unexplained".

121 **Response:** Done.

122 Line 289: Replace "decrease down" by "decreases down".

123 **Response:** Done.

124 Line 292: Replace "differences were observed" by "difference was observed".

125 **Response:** Done.

126 Line 300: Replace "other two" by "the other two".

127 **Response:** Done.

128 Line 301: "Li et al., 2013" is missing in the Reference list; there is "Li et al., 2014" in that list

129 to which not is referred within the text.

130 **Response:** Thanks. We revise the "Li et al., 2013" into "Li et al., 2014" in line 304, and the

131 "Li et al., 2014" within the text is referred in the reference list.

132 Line 339: Replace "MTLs observations" by "MTLs observation".

133 **Response:** Done.

134 Line 342: Replace "indicated these" by "indicates these".

135 **Response:** Done.

136 Line 425: Replace "this SOC compounds" by "these SOC compounds".

137 **Response:** Done.

138 Line 428: Replace "heterogonous reactions" by "heterogeneous reactions".

139 **Response:** Done.

140 Line 441: Replace "found that a" by "found a".

141 **Response:** Done.

142 Lines 728-729: There is no reference made to "Zhu et al., 2016b" within the text.

143 **Response:** The reference for "Zhu et al., 2016b" within the text was in line 66-69 in the

144 revised text "BVOCs consist primarily of isoprene, monoterpenes, sesquiterpenes, and their

145 oxygenated hydrocarbons such as alcohols, aldehydes, and ketones (Ehn et al., 2014;

146 Guenther et al., 2006) and account for the majority of the global VOC inventory (Heald et al.,

147 2008; Zhu et al., 2016a, b).".

148 **For the Supplement:**

149 Page 2, lines 1, 2, and 11: Replace "analyzed" by "measured".

150 **Response:** Done.

151 Page 2, line 12: It is unclear what is meant by "both analyses".

152 **Response:** Thanks. We revise this into "both analyses in this study and Kleindienst et al.

153 (2007)".

154 Page 6, line 2: Replace "up panel" by "upper panel" and replace "below panel" by "lower

155 panel".

156 **Response:** Done.

157 Page 7, line 2: Replace "up panel" by "upper panel".

158 **Response:** Done.

159 Page 7, line 3: Replace "below panel" by "lower panel".

160 **Response:** Done.

161 Page 8, line 2: Replace "analyzed" by "measured".

162 **Response:** Done.

163 Page 9, line 1: Replace "primary, secondary" by "primary and secondary".

164 **Response:** Done.

165 Page 9, line 2: Replace "gaactosan" by "galactosan".

166 **Response:** Done.

Tracer-based investigation of organic aerosols in marine atmospheres from marginal seas of China to the northwest Pacific Ocean

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Abstract. We investigated the geographic distributions of organic tracers in total suspended particles over marginal seas of China, including the Yellow and Bohai seas (YBS) and the South China Sea (SCS), and the northwest Pacific Ocean (NWPO) in spring, when Asian outflows strongly affect downwind marine atmospheres. The comparison of levoglucosan observed in this study with values from the literature showed that the concentrations of biomass burning aerosols over the NWPO increased largely in 2014. More observations together with ~~the~~our snapshot measurement, however, need to confirm whether the large increase occurred continuously through the last decades. The increase led to ~~the~~a mean value of levoglucosan (8.2 ± 14 ng/m³) observed over the NWPO close to that over the SCS (9.6 ± 8.6 ng/m³) and almost half of that over the YBS (21 ± 11 ng/m³). Small geographic differences in monoterpene-derived and sesquiterpene-derived secondary organic tracer concentrations were obtained among the three atmospheres, although the causes may differ. By contrast, a large difference in isoprene-derived secondary organic tracer concentrations was observed among the three atmospheres, with the sum of tracer concentrations over the SCS (45 ± 54 ng/m³) several times and approximately one order of magnitude greater than that over the YBS (15 ± 16 ng/m³) and the NWPO (2.3 ± 1.6 ng/m³), respectively. The geographic distribution of aromatic-derived secondary organic tracers was similar to that of isoprene-derived secondary organic tracers, with a slightly narrower difference, i.e., 1.8 ± 1.7 ng/m³, 1.1 ± 1.4 ng/m³ and 0.3 ± 0.5 ng/m³ over the SCS, the YBS and the NWPO, respectively. We discussed the causes of the distinctive geographic distributions of these tracers and present the tracer-based estimation of organic carbon.

1 Introduction

Aerosols that emanate from biomass burning (BB) consist primarily of carbonaceous components and inorganic salts, which can affect the climate directly by absorbing solar radiation or indirectly by acting as either cloud condensation nuclei (CCN) or ice nuclei (IN) (Bougiatioti et al., 2016; Chen et al., 2017; Hsiao et al., 2016). High BB aerosol emissions zones include boreal forests (e.g., in Eurasia and North America), tropical forests (e.g., in southeast Asia and the tropical Americas), and agriculture areas where crop residuals are burned (e.g., in developing countries such as China and India, etc.) (van der Werf et al., 2006). BB aerosols can undergo

39 long-range transport in the atmosphere, which can carry them from the continents to the oceans (Ding et al.,
40 2013; Fu et al., 2011; Kanakidou et al., 2005). For example, BB aerosols from boreal forest wildfires in Russia
41 and China reportedly made an appreciable contribution to atmospheric particle loads observed over the Arctic
42 Ocean and northwestern Pacific Ocean (NWPO) based on specific tracers of BB (Ding et al., 2013). Although
43 open wildfires from forests occur sporadically in terms of strength and occurrence frequency, global warming
44 could be conducive to vegetation fires (Running, 2006) and thus increase emissions of BB aerosols. In this
45 century, nine years were among the ten hottest global years on record, with 2014–2018 being ranked as the top
46 five hottest years (<https://www.climatecentral.org/gallery/graphics/the-10-hottest-global-years-on-record>). The
47 question is automatically raised: how do BB aerosols in the marine atmosphere in the hottest global years
48 change against those observations previously reported?

49 In addition to BB aerosols, secondary oxidation of biogenic volatile organic compounds (BVOCs) and
50 anthropogenic VOCs (AVOCs) also contribute to the particulate carbonaceous components of marine
51 atmospheres (Kanakidou et al., 2005).~~Many field and modeling studies have proposed that secondary organic~~
52 ~~aerosols (SOAs) arising from the oxidation of phytoplankton-derived isoprene may affect the chemical~~
53 ~~composition of marine atmospheric aerosols and consequently impact CCN loading and cloud droplet number~~
54 ~~concentrations (Ekström et al., 2009; Meskhidze and Nenes, 2006; Claeys et al., 2004). Secondary organic~~
55 ~~aerosols (SOAs) arising from the oxidation of phytoplankton-derived isoprene have been argued to affect the~~
56 ~~chemical composition of marine atmospheric aerosols and consequently impact CCN loading and cloud droplet~~
57 ~~number concentrations (Ekström et al., 2009; Meskhidze and Nenes, 2006), but the importance of the marine~~
58 ~~isoprene-derived SOA is still debated (Arnold et al., 2009; Claeys et al., 2010; Gantt et al., 2009; Guenther et al.,~~
59 ~~1995). For example, Gantt et al. (2009) estimated the contribution of marine isoprene-derived SOA to the OC in~~
60 ~~marine atmospheric particles ranged from <0.2% on a global scale, but to as high as 50% (sub-micron OC) over~~
61 ~~the vast regions of the oceans during the midday hours when isoprene emissions are highest.~~ Several modeling
62 studies have shown that the NWPO may experience the greatest increases in sea surface temperature and CO₂
63 input under a future warming climate ~~in the future~~ (John et al., 2015; Lauvset et al., 2017). The Kuroshio
64 Extension current system leads the NWPO to be an active subtropical cyclone basin, promoting biogenic
65 activities (Hu et al., 2018). From the perspective of global change, it is a long-term need to study the dynamic
66 changes in atmospheric aerosols derived from marine sources over the NWPO and adjacent marginal seas of
67 China, as well as their potential effects on climate.

68 ~~More importantly, BVOCs emitted from continental ecosystems and their oxidation products can significantly~~
69 ~~affect the atmosphere in remote marine areas through long-range transport (Ding et al., 2013; Fu et al., 2011; Hu~~
70 ~~et al., 2013a; Kang et al., 2018; Kawamura et al., 2017). Not limited by phytoplankton-derived isoprene,~~
71 ~~BVOCs emitted from continental ecosystems and their oxidation products can also affect the atmosphere in~~
72 ~~remote marine areas through long-range transport (Hu et al., 2013a; Ding et al., 2013; Kang et al., 2018; Fu et~~
73 ~~al., 2011; Kawamura et al., 2017). BVOCs consist primarily of isoprene, monoterpenes, sesquiterpenes, and~~
74 ~~their oxygenated hydrocarbons such as alcohols, aldehydes, and ketones (Ehn et al., 2014; Guenther et al., 2006;~~
75 ~~Ehn et al., 2014) and account for the majority of the global VOC inventory (Zhu et al., 2016a, b; Heald et al.,~~
76 ~~2008; Zhu et al., 2016a, b). However, emissions fluxes and oxidation processes of BVOCs show great variation,~~
77 ~~depending on global warming and other factors such as regional landscape, other pollutants in the ambient air,~~
78 ~~etc. (Ait-Helal et al., 2014; Claeys et al., 2004; Hu and Yu, 2013; Peñuelas and Staudt, 2010). Unlike a potential~~
79 ~~increase in BVOC-derived organics aerosols in marine atmospheres under global warming, anthropogenic~~
80 ~~VOCs and carbonaceous particles over the continents have been decreased because of effective mitigation of air~~
81 ~~pollutants in the last decades (Li et al., 2019; Murphy et al., 2011; Sharma et al., 2004; Murphy et al., 2011;~~
82 ~~Zhang et al., 2012). In the northern hemisphere, marine atmospheres are also usually affected by anthropogenic~~

83 | pollutants to some extent, most of which are derived from long-range transport from continents ([Bao et al., 2018](#);
84 | Kang et al., 2019; ~~Bao et al., 2018~~; Zhang et al., 2017). The reverse trends in BVOC and anthropogenic VOC
85 | would change the composition, sources of carbonaceous particles in marine atmospheres. Updated observations
86 | are thereby needed to reveal the change and service the future study of the impacts.

87 | In this study, we ~~analyzed-determined~~ the concentrations of some typical organic tracers in aerosol samples
88 | obtained from three cruise campaigns from the marginal seas of China, including in the South China Sea (SCS)
89 | in 2017, Yellow Sea and Bohai Sea (YBS), to the NWPO in 2014, both in springtime. We investigated the
90 | influences of BB aerosols from continents over three marine atmospheres, quantified the contributions of
91 | various precursors to the observed SOA in marine atmospheres using organic tracers established in the literature,
92 | and explored the formation pathways of SOA from their precursors during long-range transport in these hottest
93 | global years. Particularly, we conducted a comprehensive comparison of this observation with those reported in
94 | literature in terms of long-term variations and geographic distributions of these tracers, etc.

95 | **2 Materials and Methods**

96 | Total suspended particulate (TSP) samples were collected over the NWPO from 19 March to 21 April 2014,
97 | over the YBS from 30 April to 17 May 2014, and over the SCS from 29 March to 4 May 2017. All samples were
98 | collected on the upper deck of the R/V Dong Fang Hong II, which sits ~8 m above the sea surface. To avoid
99 | contamination from the ship's exhaust, samples were collected only when the ship was sailing, and the wind
100 | direction ranged from -90° to 90° relative to the bow. TSP samples were collected on quartz fiber filters
101 | (Whatman QM-A) that had been pre-baked for 4 h at 500°C prior to sampling using a high-volume sampler
102 | (KC-1000, Qingdao Laoshan Electric Inc., China). The sampling duration was 15–20 h at a flow rate of ~1000 L
103 | /min. After sampling, the sample filters were wrapped in baked aluminum foil and sealed in polyethylene bags,
104 | then stored at -20°C and transported to the laboratory. Field blanks were collected during each sampling period.
105 | However, one sampler was out of service during the cruise on the SCS. As a compromise, cellulose filters
106 | (Whatman 41) previously intended for elemental analyses were used for analyses of the organic tracers in TSP.

107 | The method for determining the concentrations of tracers was adapted from Kleindienst et al. (2007) and Feng et
108 | al. (2013). Briefly, 20 mL dichloromethane/methanol (1:1, v/v) was used for ultrasonic extraction of 40 cm² of
109 | each filter at room temperature three times. The combined extracts were filtered, dried under a gentle stream of
110 | ultrapure nitrogen, and then derivatized with 100 μL N,O-bis-(trimethylsilyl)-trifluoroacetamide (BSTFA,
111 | containing 1% trimethylchlorosilane as a catalyst) and 20 μL pyridine at 75°C for 45 min. Gas chromatography
112 | mass spectrometry (GC-MS) analyses were conducted with an Agilent 6890 GC/5975 MSD. Prior to solvent
113 | extraction, methyl-β-D-xylanopyranoside (MXP) was spiked into the samples as an internal/recovery standard.
114 | Hexamethylbenzene was added prior to injection as an internal standard to check the recovery of the surrogates.

115 | Like those reported by Feng et al. (2013), the primary organic tracers analyzed in this study included
116 | levoglucosan (LEVO), mannosan, and galactosan. Four types of secondary organic tracers were used:
117 | isoprene-derived secondary organic tracers (SOA_I) including 2-methylglyceric acid (2-MGA), C5-alkene triols
118 | (cis-2-methyl-1,3,4-trihydroxy-1-butene, 3-methyl-2,3,4-trihydroxy-1-butene and
119 | trans-2-methyl-1,3,4-trihydroxy-1-butene), and MTLs (2-methylthreitol and 2-methylerythritol);
120 | monoterpene-derived secondary organic tracers (SOA_M) including 3-hydroxyglutaric acid (HGA),
121 | 3-hydroxy-4,4-dimethylglutaric acid (HDMGA), and 3-methyl-1,2,3-butanetricarboxylic acid (MTBCA); the
122 | sesquiterpene-derived secondary organic tracer (SOA_S) β-caryophyllinic acid; and the aromatic
123 | (toluene)-derived secondary organic tracer (SOA_A) 2,3-dihydroxy-4-oxopentanoic acid (DHOPA). LEVO was
124 | quantified based on authentic standards in this study. While the SOA tracers without available commercial
125 | standards were quantified using methyl-β-D-xylanopyranoside (MXP) as a surrogate. To reduce the uncertainty

126 of quantification, relative response factors of the target tracers to MXP were estimated by comparing the area
127 ratio of typical target ions to MXP to that of total ions in selected samples that showed high concentrations of
128 the target tracers (Feng et al., 2013).

129 Field blanks and laboratory blanks (~~run-ran~~ every 10 samples) were extracted and analyzed in the same manner
130 as the ambient samples. Target compounds were nearly always below the detection limit in field and laboratory
131 blanks. Recoveries of the surrogate (MXP) were in the range of 70–110%. The reported results were corrected
132 for recovery, assuming that the target compounds had the same recovery as the surrogate. Duplicate analyses
133 indicated that the deviation was less than 15%.

134 However, the substitution of cellulose filters (Whatman 41) during the cruise on the SCS led to increased field
135 blank values for some tracers. The tracer concentrations in those samples were, however, over three times higher
136 than the field blank values, except for those of mannosan and galactosan. Data for mannosan and galactosan
137 were thus not available, nor were the total organic carbon concentrations, for samples collected during the cruise
138 on the SCS.

139 The concentrations of organic carbon (OC) and element carbon (EC) in each sample were analyzed with a DRI
140 2001A thermal/optical carbon analyzer (Atmoslytic Inc., Calabasas, CA, USA) using the IMPROVE
141 temperature program (Wang et al., 2015). All filters before and after sampling were weighted at a glovebox
142 under ~~the~~ controlled ambient temperature and relative humidity. Mass concentrations of TSP, however, should
143 be treated as semi-quantitative results by considering analytic errors of quartz fiber filters (Yao et al., 2009).

144 3. Results and Discussion

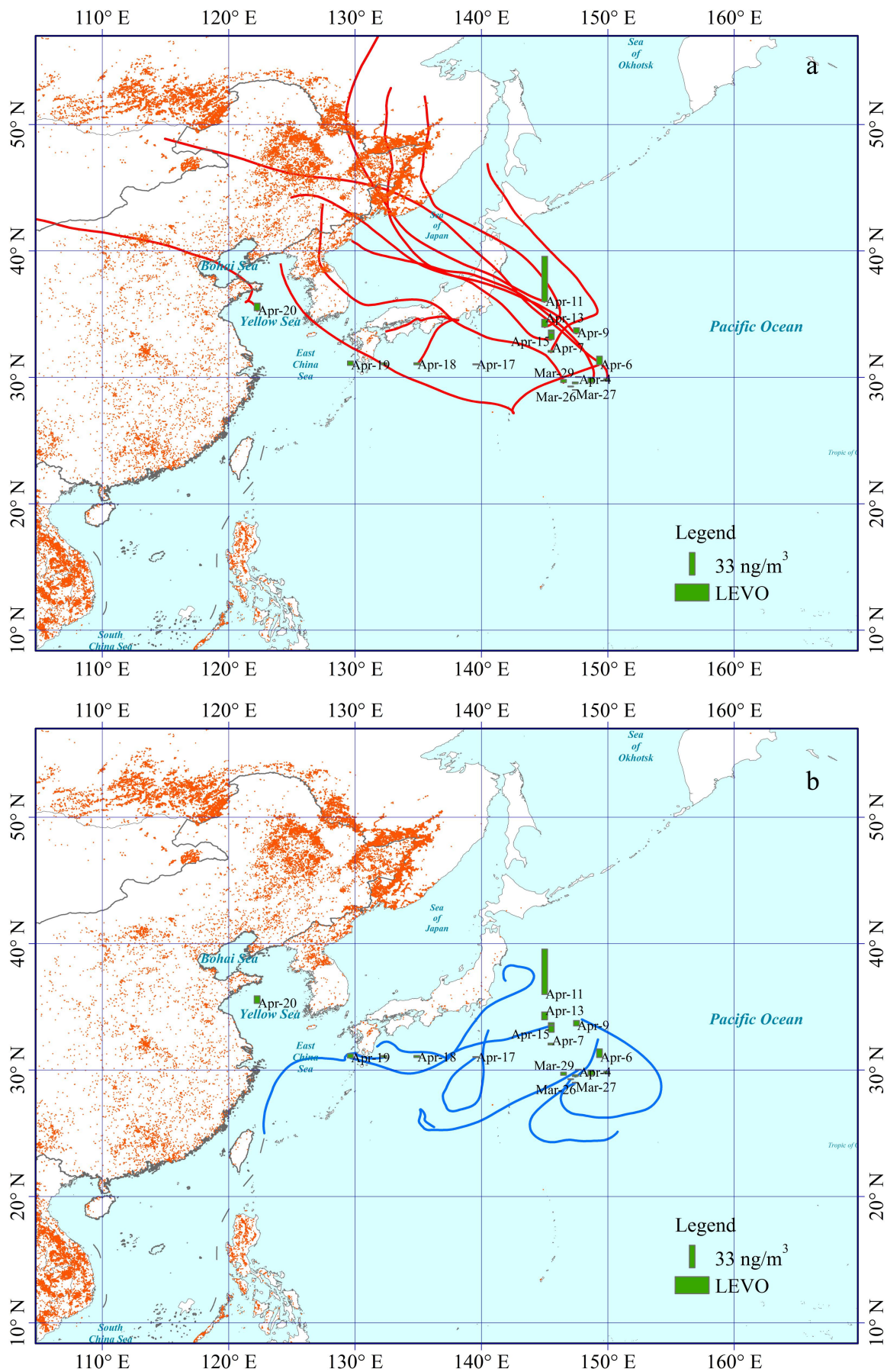
145 3.1 Spatiotemporal distributions of LEVO

146 Levoglucosan, mannosan, and galactosan produced by the pyrolysis of cellulose and hemicellulose have been
147 widely used as organic tracers of BB aerosols in ambient air (Ding et al., 2013; Fu et al., 2011; Feng et al.,
148 2013). The mean levels of LEVO in TSP collected during the cruises on the NWPO and the SCS were
149 comparable, at 8.2 ng/m³ and 9.6 ng/m³, respectively (Figure S1, Table 1). They were almost half of the mean
150 value of 21 ng/m³ during the cruise on the YBS, where high concentrations of BB aerosols have been observed
151 in continental atmospheres upwind of the YBS mainly from wildfires and the burning of crop residue, wildfire,
152 etc. (Feng et al., 2012; Feng et al., 2013; Yang et al., 2014; ~~Feng et al., 2012; Feng et al., 2013~~). Unlike the
153 smaller difference among the means values, the concentration of LEVO fluctuated greatly among TSP samples
154 in each oceanic zone, ranging from 0.5 to 65 ng/m³ over the NWPO, from 1.0 to 30 ng/m³ over the SCS and
155 from 2.5 to 42 ng/m³ over the YBS (Fig. S1). High spatiotemporal variation in LEVO in TSP has also been
156 observed in literature, with concentrations of LEVO fluctuating around 0.2–41 ng/m³ during Arctic to Antarctic
157 cruises from July to September 2008 and from November 2009 to April 2010 (Hu et al., 2013b). Hu et al.
158 (2013b) also reported the highest LEVO concentrations occurring at mid-latitudes (30°–60° N and S) and the
159 lowest at Antarctic and equatorial latitudes over the several months of sampling. This distinctive geographical
160 distribution was not observed in the present study, as there were no significant differences in LEVO in TSP
161 between the SCS and NWPO ($P > 0.05$).

162 Narrow spatiotemporal variation in LEVO in TSP has been reported during summer sampling over the North
163 Pacific Ocean and the Arctic in 2003, with maximum and mean values as low as 2.1 ng/m³ and 0.5 ng/m³,
164 respectively (Ding et al., 2013). A lower mean value of LEVO of 1.0 ng/m³ has also been reported in the spring
165 over the island of Chichi-jima from 2001 to 2004 (Mochida et al., 2010), while the levels increased to 3.1 ± 3.7
166 ng/ m³ in TSP collected on the island of Okinawa in 2009–2012 (Zhu et al., 2015). ~~Using these previous~~

167 | observations as a reference (Table 1), our observations suggested that the BB aerosols from the long-range
168 | transport over the NWPO in 2014 largely increased. Thus, an important question is raised, i.e., does the increase
169 | occur continuously and largely over the last decades in marine atmospheres over the NWPO? Due to the lack of
170 | BB sources in oceans, large spatiotemporal variation in the concentrations of LEVO in the marine atmosphere
171 | may be related to the long-range transport of atmospheric particles from continents. Thus, 72 h back trajectories
172 | of air masses at a height of 1000 m during our sampling periods (Figs. 1, 2) were calculated using the HYSPLIT
173 | model (<https://ready.arl.noaa.gov/HYSPLIT>). Based on the calculated back trajectories, TSP samples could be
174 | classified into two categories with Category 1 representing continent-derived aerosol samples and Category 2
175 | being ocean-derived aerosol samples. All 12 samples collected over the YBS fell into Category 1 (Fig. 2). Half
176 | (11/19) of the samples collected over the NWPO were classified into Category 1 (Fig. 1). A significant
177 | difference ($p < 0.05$) was obtained between the concentrations of LEVO in Category 1 ($13 \pm 18 \text{ ng/m}^3$) and
178 | Category 2 ($2.0 \pm 1.8 \text{ ng/m}^3$) over the NWPO. The values in Category 2 were closer to the springtime
179 | observations reported by Mochida et al. (2010) and Zhu et al. (2015) as well as the summer observations
180 | reported by Ding et al. (2013), reflecting the marine background value less affected by continental air masses.
181 | On the other hand, the much higher values in Category 1 than Category 2 further indicated a large increase in
182 | contribution of BB aerosols being transported from the continents to the remote marine atmosphere in 2014.
183 | On 11 April 2014 over the NWPO, an episode of high LEVO concentration of 65 ng/m^3 occurred (Fig. 1). Like
184 | LEVO, the concentrations of galactosan and mannosan in the sample were also the highest among all samples
185 | collected over the NWPO. This sample was collected in the oceanic zone, approximately 500 km from the
186 | continent of Japan. A combination of air mass back ~~trajectories~~traeries and NASA's FIRMS Fire Map indicated
187 | strong BB aerosol emissions from intense fire events in Siberia, followed by long-range transport with the
188 | westerly wind as the major contributors to this anomaly (Fig. 1). A similar episodic concentration of LEVO of
189 | 27 ng/m^3 in TSP was observed once previously over the NWPO during a circumnavigation cruise (Fu et al.,
190 | 2011). By combining satellite data with other observations, many studies in literature have found that BB
191 | aerosols from major forest fires and smoke events in Siberia could be transported downwind to remote marine
192 | regions not only in spring, but also in summer (~~Generoso et al., 2007~~; Ding et al., 2013; [Generoso et al., 2007](#);
193 | Huang et al., 2009). In a few cases, BB aerosols have been reported to have reached as far as the adjacent Arctic
194 | region (~~Warneke et al., 2010~~[Generoso et al., 2007](#); ~~Warneke et al., 2010~~[Generoso et al., 2007](#)). Van der Werf
195 | et al. (2006) estimated the emissions of BB aerosols from Eurasia to be much larger than those from North
196 | America. Thus, it is not surprising that the concentrations of LEVO over the NWPO were much higher than
197 | those over the eastern North Pacific and western North Atlantic at similar latitudes (Hu et al., 2013b).
198 | In addition, both galactosan and mannosan showed strong linear correlations with LEVO ($R^2 = 0.98$, $p < 0.05$)
199 | in TSP collected over the NWPO and YBS in this study. These strong correlations indicate that the three tracers
200 | were probably derived from the same BB sources. Previous studies have reported LEVO/mannosan (L/M) ratios
201 | of 3–10, 15–25, and 25–40 from softwood, hardwood, and crop-residue burning, respectively (Kang et al., 2018;
202 | Zhu et al., 2015). The calculated L/M ratios in TSP collected over the NWPO were 19 ± 4 in this study, which
203 | implies dominant contributions from herbaceous plants and hardwood. The calculated L/M ratios in TSP
204 | collected over the YBS were 14 ± 11 , indicating mixed sources.
205 | In all, 5 of 13 samples collected over the SCS were classified into Category 1, with air masses identified as
206 | originating from either the continental areas of South China or the Philippines (Fig. 2). The concentration of
207 | LEVO fluctuated around $17 \pm 12 \text{ ng/m}^3$ in Category 1 but decreased to $3.6 \pm 3.4 \text{ ng/m}^3$ in Category 2. However, no
208 | significant differences ~~were was~~ found between categories due to the large variation in LEVO concentration
209 | among the ~~limited number of samples~~limited samples in Category 1 ($p > 0.05$). Forest fires occur accidentally,
210 | leading to the large variation in LEVO in Category 1. Southern Asia has been reported to be one of the greatest

211 emissions sources of BB aerosols worldwide (van der Werf et al., 2006), which likely led to the higher mean
212 value of LEVO in Category 1. However, the LEVO level observed over the SCS in Category 2 was closer to that
213 reported from low-latitude regions (2.7 ± 1.1 ng/m³, Table 1) collected during a global circumnavigation cruise
214 (Hu et al., 2013b). Hu et al. (2013b) argued that their low observed concentrations may have been associated
215 with intense wet deposition, degradation as well as intensive moist convection that occurred in the tropical
216 region during their summer cruise. Unfortunately, no previous observations of LEVO in spring can allow us
217 analyzing the long-term variation in contribution of BB aerosols therein. However, this observation can be used
218 for future comparison.



219

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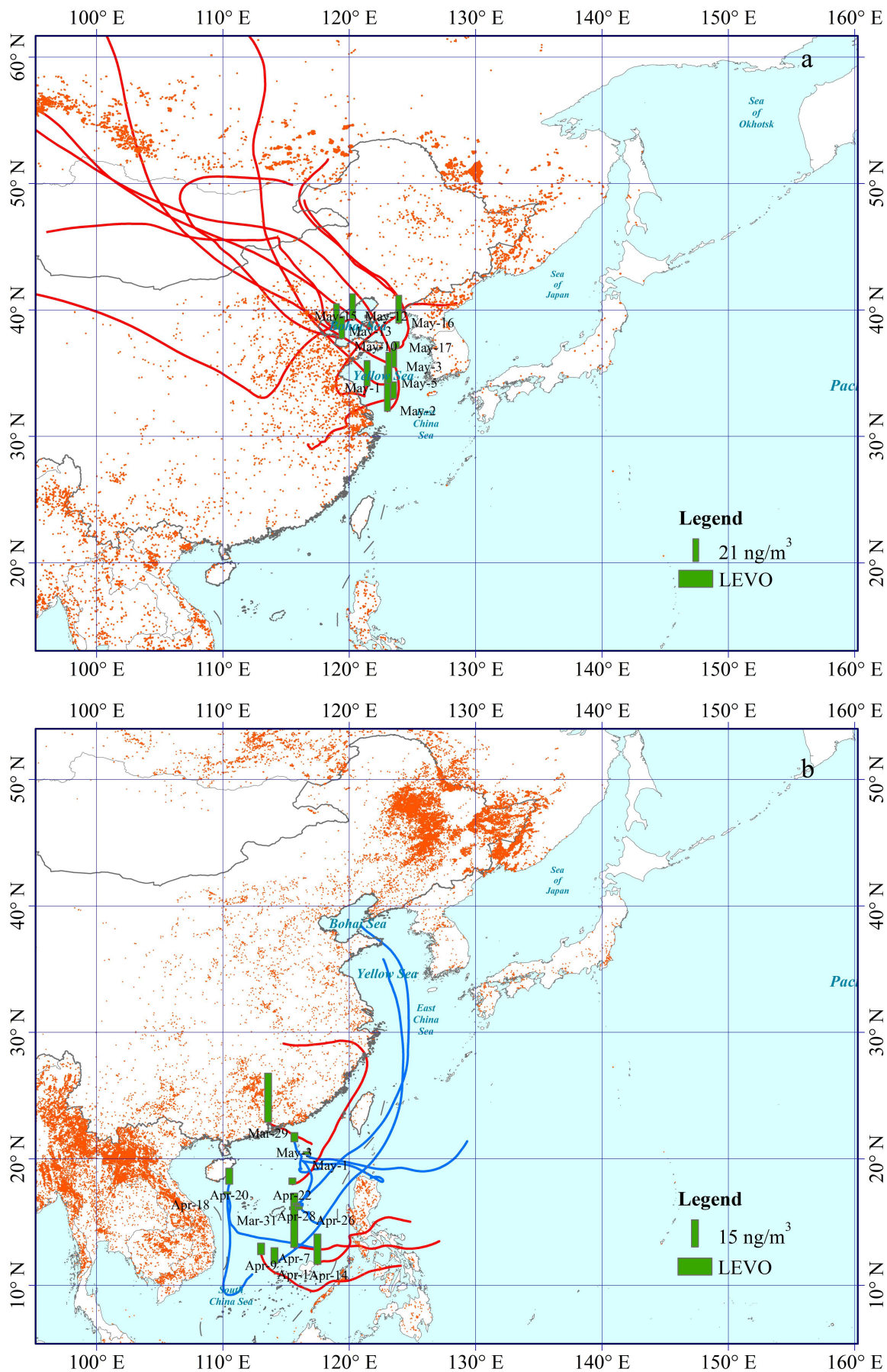
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223

Figure 1. Spatial distribution of LEVO in TSP over the NWPO in spring of 2014 and 72-hrs back trajectory associated with each TSP sample. The red lines represent that air masses can be derived from the continent (a, Category 1); the blue lines represent that air masses may be derived mainly from the oceans (b, Category 2). The red dots represent the locations of fires from Fire Information for Resource

224 Management System (FIRMS, <https://firms.modaps.eosdis.nasa.gov/>). And the base map was from
225 Resource and Environment Data Cloud 210 Platform, DOI: 10.12078/2018110201.

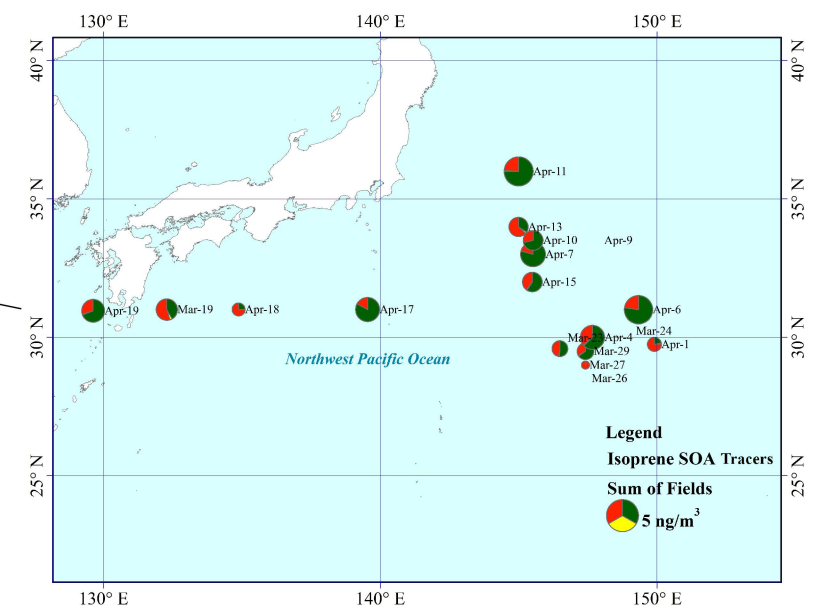
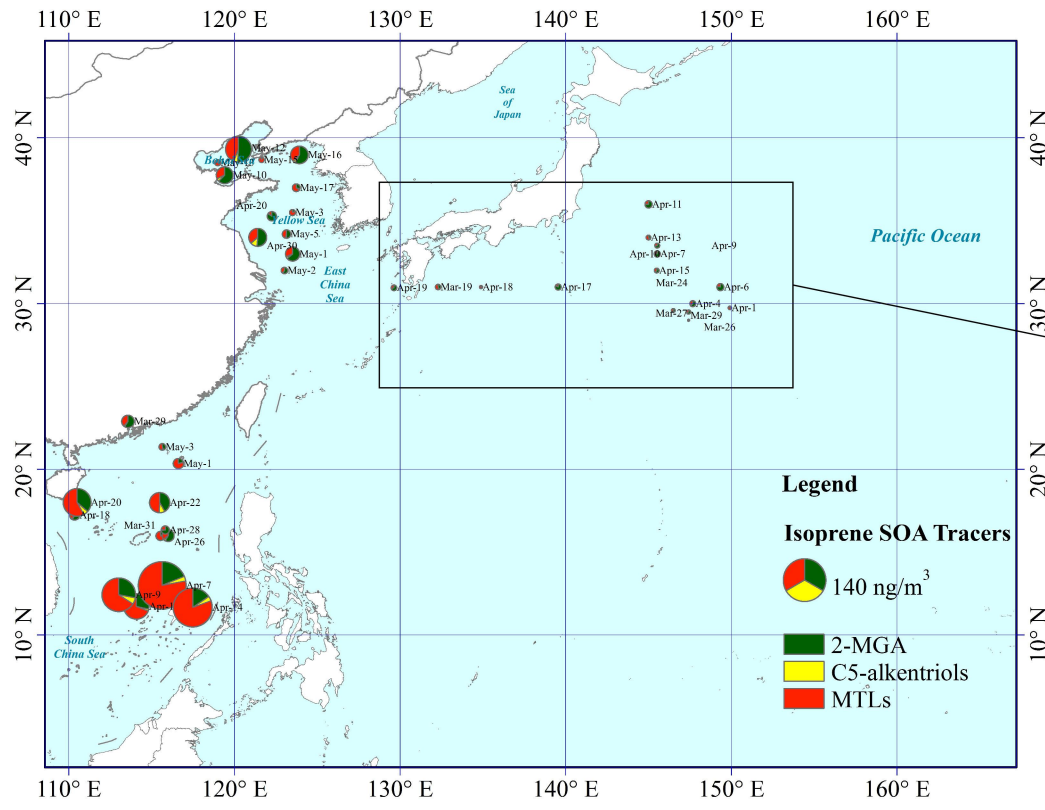


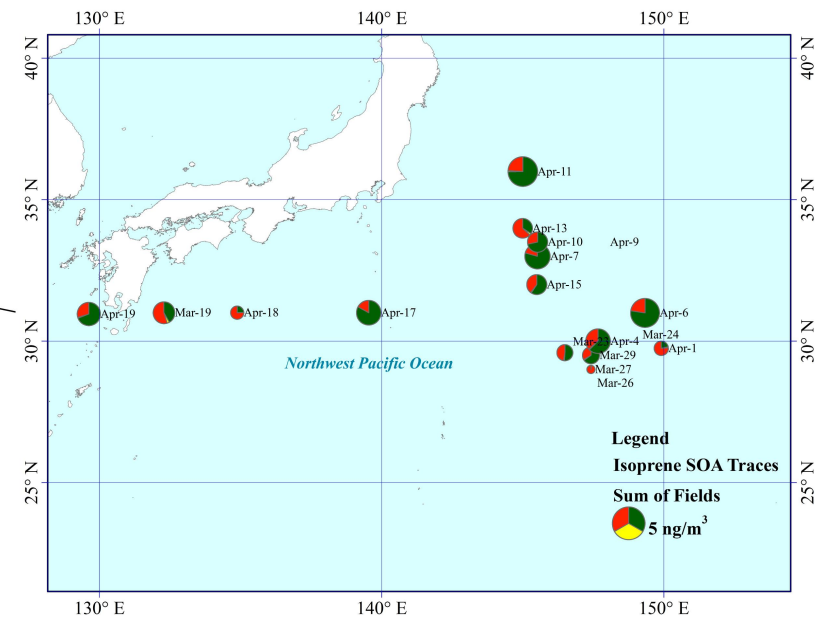
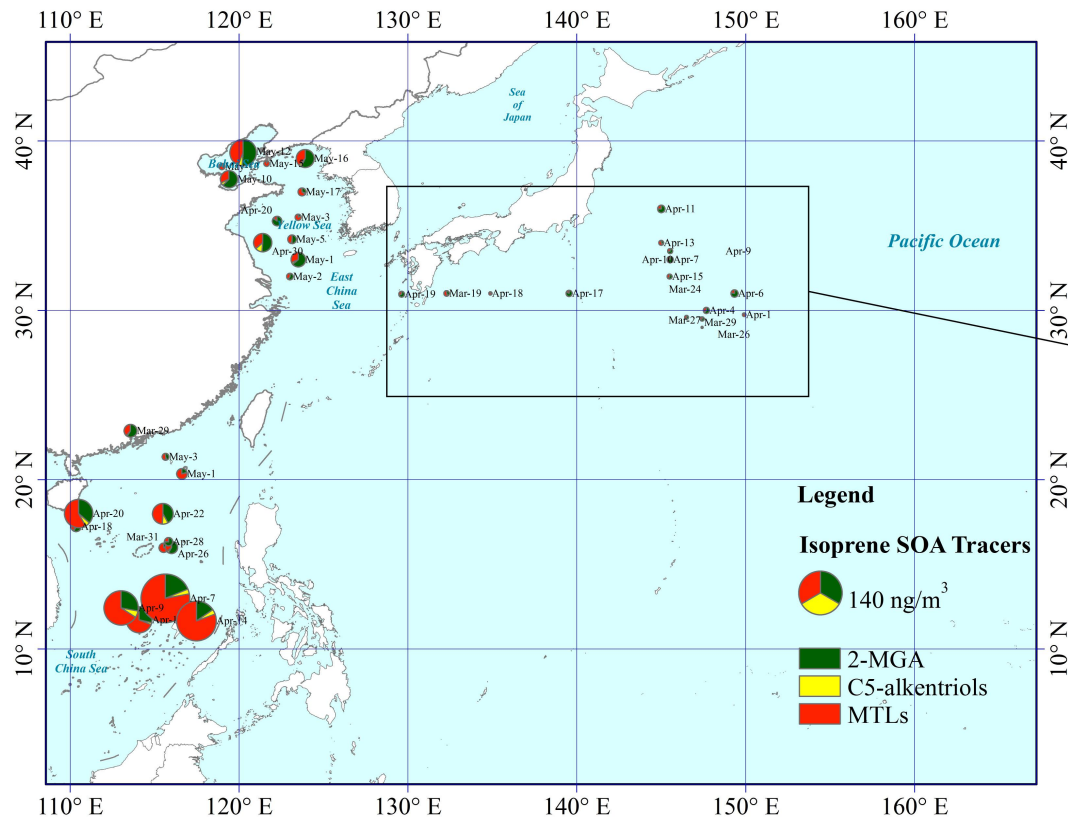
226
227 **Figure 2. Spatial distribution of LEVO over the YBS (a, 2014), and SCS (b, 2017), detailed information**
228 **described in Figure 1. And the base map was from Resource and Environment Data Cloud 210 Platform,**

230 3.2 Spatiotemporal distributions of SOA_I tracers

231 SOA_I tracers were detected during all three cruises. The sum of SOA_I tracers showed a decreasing trend of up to
232 approximately one order of magnitude from marginal seas to the open ocean, i.e., 45 ± 54 ng/m³ in TSP over the
233 SCS, 15 ± 16 ng/m³ over the YBS and 2.3 ± 1.6 ng/m³ over the NWPO (Fig. S1). The highest sum value of SOA_I
234 tracers over the SCS was 176 ng/m³, indicating strong photochemical formation of SOA from biogenic volatile
235 organics (Fig. 3). The geographical distribution of SOA_I tracers in this study was generally consistent with those
236 reported by Hu et al. (2013a), with higher concentrations of these tracers in atmospheric particles collected from
237 low-latitude oceanic zones (30° S–30° N) due to large emissions from tropical forests and strong photochemical
238 reactions. Their reported average contents of SOA_I tracers in low-latitude oceanic zones fluctuated around
239 9.2 ± 6.7 ng/m³, much lower than those measured in this study.

240 When the sum of SOA_I tracers in each sample was examined separately according to the air mass source, a
241 significant difference was found over the SCS between Category 1 (85 ± 66 ng/m³) and Category 2 (19 ± 22
242 ng/m³), with significance at $p < 0.01$. The average contribution of SOA_ISOA tracers to TSP mass concentration
243 over the SYS was higher in category 1 ($0.4\% \pm 0.6\%$) than in category 2 ($0.06\% \pm 0.07\%$). The tracer values
244 were 2.7 ± 1.8 ng/m³ in Category 1 and 1.7 ± 1.0 ng/m³ in Category 2 over the NWPO, where no significant
245 difference between the two categories was found ($p > 0.05$). The average contribution of SOA_ISOA tracers to
246 TSP mass concentration over the NWPO was higher in category 1 ($0.008\% \pm 0.005\%$) than that in category 2
247 ($0.005\% \pm 0.005\%$). Supposed that concentrations of the tracers in Category 2 were completely contributed by
248 marine sources, it can be inferred that SOA_I carried by continental air masses increased sharply over the SCS.
249 However, it was not the case over the NWPO. Because all samples over the YBS fell into Category 1, this
250 comparison could not be made for the YBS.





252
 253 **Figure 3. Spatial distribution of SOA₁ tracer compounds over three marine regions, ECS-YBS and NWPO in 2014, SCS in 2017. The area of the pie indicates the concentration of**
 254 **total SOA₁ tracers. The base map was from Resource and Environment Data Cloud Platform, DOI: 10.12078/2018110201.**

255 3.3 Spatiotemporal distributions of SOA_M, SOA_s tracers

256 The sum of SOA_M tracers including HGA, HD-MGA, and MBTCA was greatest over the SCS region (3.5±6.0
257 ng/m³), where the concentration was approximately double that over the YBS (1.6±2.0 ng/m³) and NWPO
258 regions (1.6±2.7 ng/m³) (Fig. S1), but no significant differences were identified between any two campaigns.
259 The concentrations of SOA_M tracers were almost one magnitude lower than those of SOA_I tracers. Due to the
260 unique contribution of terpene-derived SOA to nucleation and growth of newly formed particles in the
261 atmospheres (Ehn et al., 2014; Gordon et al., 2017; Zhu et al., 2019; Ehn et al., 2014), the SOA_M may primarily
262 cause indirect climate effects rather than direct effects of aerosols in the marine atmosphere. The difference in
263 mean SOA_M concentration between the SCS and NWPO narrowed to a factor of two, in contrast to the
264 differences of approximately one order of magnitude in mean SOA_I between the two types of atmospheres. The
265 precursors of SOA_M tracers derive mainly from coniferous forests (Duhl et al., 2008) and the decreasing
266 proportion of coniferous forests in subtropical and tropical regions may partially explain the smaller spatial
267 difference in SOA_M tracers over the SCS compared to the YBS and NWPO. However, the comparable SOA_M
268 levels over the YBS and NWPO have not yet been explained.

269 Only three SOA_M tracers were measured in this study, but other SOA_M tracers have been measured and reported
270 in marine atmospheres (Fu et al., 2011; Kang et al., 2018; Fu et al., 2011). In order to compare our results with
271 the total amount of SOA_M tracers in the literature, the total amounts measured in this study were multiplied by a
272 factor of 3.1 (described in supporting information Sect. S1, Fig. S4) according to the chamber results obtained
273 by Kleindienst et al. (2007). The adjusted values over the SCS were closer to the mean of 11.6 ng/m³ observed
274 over the [East China Sea \(ECS\)](#) (Kang et al., 2018) and the lower values of 9.80–49.0 ng/m³ observed among 12
275 continental sites in China (Ding et al., 2016). The adjusted total amounts of SOA_M over the NWPO and YBS
276 were comparable to previous observations of 3.0±5.0 ng/m³ collected from the Arctic to Antarctic in 2008-2010
277 (Hu et al., 2013a), but much higher than observations of 63±49 pg/m³ over the North Pacific and Arctic in 2003
278 (Ding et al., 2013). This may also imply a substantial increase in SOA_M in the last decades, although more
279 investigations are needed to confirm [this](#).

280 β-Caryophyllene is a major sesquiterpene emitted from plants such as Scots pine and European birch (Duhl et al.,
281 2008; Tarvainen et al., 2005). β-Caryophyllinic acid is formed through the ozonolysis or photo-oxidation of
282 β-caryophyllene. The highest levels of β-caryophyllinic acid were observed over the YBS (0.13±0.03 ng/m³),
283 followed by the SCS (0.08±0.11 ng/m³) and NWPO (0.05±0.09 ng/m³) (Fig. S1). The spatial distribution of
284 β-caryophyllinic acid clearly did not follow the general trend of biogenic SOA, with the highest values over the
285 SCS followed by the YBS. Compared to values from the literature, our results are much higher than those over
286 the North Pacific and Arctic Oceans (2.4±5.4 pg/m³) (Ding et al., 2013) but much lower than observations over
287 the East China Sea reported by Kang et al. (2018), where β-caryophyllinic acid was reported to be in the range
288 of 0.16–17.2 ng/m³ with a mean of 2.9 ng/m³. The large differences in β-caryophyllinic acid content observed in
289 various campaigns remains unexplained.

290 3.4 Spatiotemporal distributions of SOA_A tracers

291 When the concentrations of DHOPA in TSP were examined, the highest concentrations occurred over the SCS
292 (1.8±1.7 ng/m³), followed by the YBS (1.1±1.4 ng/m³), and the lowest values were recorded in the NWPO
293 region (0.3±0.5 ng/m³) (Fig. S1). The decreasing extent of the DHOPA from the SCS to the NWPO was
294 approximately three times less than that of SOA_I tracers but approximately three times larger than that of SOA_M

295 tracers. Ding et al. (2017) reported annual averages of DHOPA among various sites in China, which ranged from
296 1.2 to 8.8 ng/m³. The concentrations of DHOPA observed over the SCS and the YBS were similar to the lower
297 values observed in upwind continental atmospheres.

298 Formation of DHOPA depends on the molecular structures of aromatics, as well as concentrations of free
299 radicals and oxidants, etc. (Henze et al., 2008; Li et al., 2016; Henze et al., 2008). The mean value of DHOPA in
300 Category 1 (0.43±0.65 ng/m³) was nearly twice that in Category 2 (0.20±0.31 ng/m³) over the NWPO (p > 0.05).
301 With two samples with high DHOPA (1.2, 2.1 ng/m³) in Category 1 to be excluded, the recalculated average
302 DHOPA decreases down to 0.17±0.21 ng/m³. The continent-derived DHOPA seemingly yielded a minor
303 contribution to the observed values over the NWPO, except during strong long-range transport episodes.
304 Similarly, the mean values of DHOPA were same in Category 1 (1.8±2.1 ng/m³) and Category 2 (1.8±1.5 ng/m³)
305 samples collected over the SCS and no significant differences were observed between two categories. Much
306 stronger UV radiation occurs over the SCS than the YBS, which may contribute to the elevated DHOPA level
307 over the SCS. Aside from continent-derived precursors, oil exploration and heavy marine traffic over the SCS
308 are also potential contributors to the higher DHOPA levels therein, and this link requires further investigation.
309 Previous field observations in China have demonstrated that biofuel or biomass combustion emissions act as
310 important sources of aromatics in the atmosphere (Zhang et al., 2016), as evidenced by the association between
311 the nationwide increase in DHOPA during the cold period and the enhancement of BB emissions (Ding et al.,
312 2017). In this study, no linear correlation was obtained between DHOPA and LEVO in samples collected over
313 the SCS and the other two campaigns, leaving emissions other than BB emissions, e.g., solvent use, oil
314 exploration, marine traffic, etc., as the major precursors for DHOPA in these marine atmospheres (Li et al.,
315 2013, 2014).

316 3.5 Causes for high photochemical yields of SOA_I over the SCS

317 Because higher concentrations of SOA_I were observed in TSP samples collected over the SCS, the composition
318 of SOA_I tracers was further investigated in terms of their formation pathways and sources. Based on the results
319 of chamber experiments, Surratt et al. (2010) proposed different formation mechanisms for 2-MGA and MTLs.
320 2-MGA is a C₄-dihydroxycarboxylic acid, which forms through a high-NO_x pathway. MTLs and C₅-alkene
321 triols are mainly products of the photooxidation of epoxydiols of isoprene under low-NO_x conditions.

322 MTLs acted as the dominant compounds among SOA_I tracers in most TSP samples collected over the SCS, with
323 concentrations of 31±42 ng/m³ (Fig. 3). The ratio of 2-MGA/MTLs ranged from 0.2 to 3.1, with a median value
324 of 0.6. The ratio exceeded the unity in only 4 of 13 samples. This result allowed us to infer that the observed
325 SOA_I tracers were generated mainly under low-NO_x conditions. Although the concentration of
326 2-methylerythritol was nearly double that of 2-methylthreitol, they were highly correlated (R² = 0.99, p < 0.05)
327 because of their shared formation pathway. Satellite data showed that the NO₂ levels in South China and the
328 Philippines were low, except in a few hotspots (Fig. S2). Such low-NO_x conditions favor the formation of
329 MTLs rather than 2-MGA over the tropical SCS. The isoprene emitted from plants growing on oceanic islands
330 may also undergo chemical conversion to SOA under low-NO_x conditions, and low-NO_x conditions are always
331 expected in remote marine atmospheres (Davis et al., 2001).

332 In general, zonally and monthly averaged OH concentrations around 15°N are ~50% were greater than those
333 around 35 °N (Bahm and Khalil, 2004). Thus, enhanced formation of MTLs is theoretically expected under the
334 strong UV radiation of tropical regions. However, no significant correlation between the concentrations of
335 MTLs and UV radiation was obtained over the SCS (data not shown) possibly due to the influences of various
336 air masses. A field study showed that MTL yields were positively correlated with ambient temperature in

337 continental atmospheres (Ding et al., 2011). 2-MGA yields, in contrast, showed no significant correlation with
338 ambient temperature in this study. Moreover, lower relative humidity may enhance the formation of 2-MGA in
339 the particulate phase but not for MTLs (Zhang et al., 2011). Variation in ambient temperature and relative
340 humidity may complicate the relationship between the concentrations of SOA_I tracers and UV radiation over the
341 SCS.

342 In addition, the MTLs concentration in Category 1 (62 ± 55 ng/m³) was larger than that in Category 2 (11 ± 14
343 ng/m³). The more abundant MTLs associated with Category 1 was most likely related to long-range transport of
344 these chemicals from upwind continental areas, the oxidation of continental precursors in the marine atmosphere,
345 or both. Large emissions of isoprene were expected from tropical forests upwind of the SCS due to the high
346 vegetation coverage and high ambient temperature of such areas (Ding et al., 2011; Rinne et al., 2002). Global
347 estimates show tropical trees to be responsible for ~80% of terpenoid emissions and ~50% of other VOC
348 emissions (Guenther et al., 2012).

349 In a clean marine atmosphere, phytoplankton is the sole source of isoprene emissions over the oceans (Bonsang
350 et al., 1992; Broadgate et al., 1997). Chlorophyll-a has been widely employed as a measure of phytoplankton
351 abundance and a proxy for predicting isoprene concentrations in water (Hackenberg et al., 2017). The
352 satellite-derived chlorophyll-a level during the study period over the SCS was below 0.45 mg/m³, excluding
353 coastal areas (Fig. S3). The MTLs observations of 11 ± 14 ng/m³ in Category 2 should be considered as the upper
354 limitation value derived from marine phytoplankton in the SCS. Although air masses differed between
355 Categories 1 and 2, a good correlation was obtained between MTLs and 2-MGA when the data in the two
356 categories was pooled for analyses ($R^2 = 0.77$, $P < 0.01$). This strong correlation indicated-indicates these tracers
357 are primarily formed through shared pathways. However, this correlation was poor over the NWPO, as
358 discussed below.

359 3.6 Origin and formation of SOA_I over the NWPO

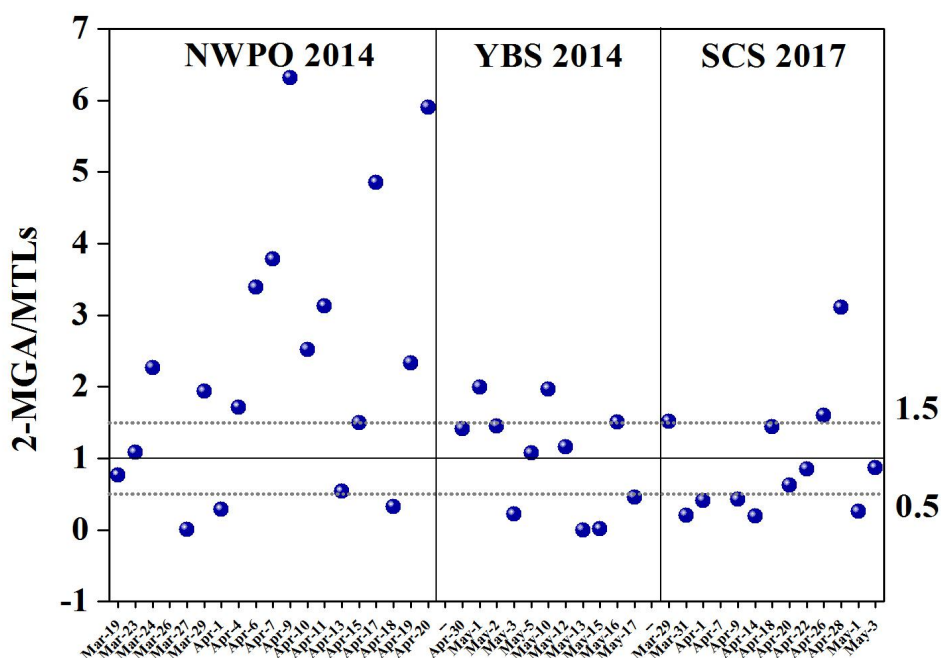
360 Over the NWPO, the concentration of 2-MGA was 1.6 ± 1.5 ng/m³, which was generally dominant among SOA_I
361 tracers, followed by MTLs (0.7 ± 0.3 ng/m³) and C5-alkene triols (0.03 ± 0.02 ng/m³). When the ratio of
362 2-MGA/MTLs was further examined, it varied greatly from <0.1 to 6.3, with a median value of 2.1. Most ratios
363 observed over the NWPO in this study were far greater than the values of 0.18–0.59 reported by Hu et al. (2013a)
364 from a global circumnavigation cruise, and also greater than 0.87–1.8 reported in urban areas of California
365 (Lewandowski et al., 2013) and the maximum value of 2.0 obtained over the YBS. Ding et al. (2013) also
366 reported ratios that fluctuated greatly from 0.5 to 10 with a median value of 3.3 during a summer cruise in the
367 NWPO and Arctic Ocean in 2003. The large 2-MGA/MTL ratios over the NWPO appeared to be highly
368 consistent over two independent sampling campaigns.

369 The compound profile of SOA_I tracers over the NWPO implied high-NO_x conditions allowing oxidation of
370 isoprene to generate the SOA_I present in most samples. Such high-NO_x conditions are impossible in a remote
371 marine atmosphere, as indicted in Figure S2. Given that the lifespan of isoprene in the atmosphere is only
372 several hours (Bonsang et al., 1992), the long-range transport of oxidation products formed under high NO_x
373 levels over the continents likely led to the 2-MGA-dominated composition of SOA_I. Based on air mass back
374 trajectories, this long-range transport may involve 2-MGA originating from Siberia, northeastern China, or
375 Japan.

376 Organic aerosols over the NWPO were strongly influenced by forest fires that take place in Siberia during
377 spring and summer almost every year (Ding et al., 2013; Huang et al., 2009). Previous emissions inventory
378 studies have reported high isoprene and NO_x emissions from various BB types (Akagi et al., 2011; Andreae and

379 Merlet, 2001). Ding et al. (2013) thus argued that an increase in emissions of isoprene in the presence of BB,
 380 followed by its chemical conversion under high-NO_x conditions, may lead to transport over thousands of
 381 kilometers and hold at the detectable concentrations in the remote marine atmosphere over the NWPO. The
 382 same argument may hold true for the elevated ratios of 2-MGA/MTLs observed over the NWPO in this study
 383 (Fig. 4). However, we did not find a significant correlation between 2-MGA and LEVO over the NWPO. The
 384 decomposition of LEVO reported in literature (Fraser and Lakshmanan, 2000; Hennigan et al., 2010; Hoffmann
 385 et al., 2010; Fraser and Lakshmanan, 2000) may lower the correlation between them. However, whether 2-MGA
 386 can decompose in ambient air remains poorly understood.

387 On the other hand, the ratios of 2-MGA/MTLs in 3 of 19 samples collected over the NWPO were below 0.5
 388 (Figure 4). In these cases, the oxidation of isoprene under low-NO_x conditions likely dominated the generation
 389 of SOA_I. The ratios of 2-MGA/MTLs were 0.5–1.5 in 4 of 19 samples, suggesting mixed contributions to SOA_I
 390 from the oxidation of isoprene under low-NO_x conditions and high-NO_x conditions. As the major formation
 391 pathways of 2-MGA and MTLs varied greatly among samples, no significant correlation ($R^2 = 0.12$, $p > 0.05$)
 392 was obtained between 2-MGA and MTLs over the NWPO. Recall that the tracer values of SOA_I were 2.7 ± 1.8
 393 ng/m³ in Category 1 and 1.7 ± 1.0 ng/m³ in Category 2. This implied that SOA_I derived from marine sources was
 394 comparable to that derived from the continent outflows.



395
 396 **Figure 4. Spatial ratio of 2-MGA/MTLs among SOA_I tracers over three marine regions.**

397 **3.7 Source apportionment of secondary organic carbon (SOC)**

398 The tracer-based approach developed by Kleindienst et al. (2007) was applied to estimate the concentrations of
 399 SOC and WSOC_{BB}, as follows:

$$[SOC] = \frac{\sum_i [tri]}{f_{soc}} \quad (1)$$

400

$$[WSOC_{BB}] = \frac{C_{tracer}}{f_{tracer/WSOC_{BB}}} \quad (2)$$

401 where $\Sigma_i(\text{tri})$ is the sum of concentrations of the selected suite of tracers for a precursor, and f_{SOC} is the mass
 402 fraction of tracer compounds in SOC generated from the precursor in chamber experiments. Assuming that the
 403 f_{SOC} values in ambient air match those in the chamber, the f_{SOC} values for precursors such as isoprene,
 404 monoterpenes, β -caryophyllene, and aromatics were $0.155 \pm 0.039 \mu\text{g}/\mu\text{gC}$, $0.231 \pm 0.111 \mu\text{g}/\mu\text{gC}$, 0.023 ± 0.0046
 405 $\mu\text{g}/\mu\text{gC}$, and $0.00797 \pm 0.0026 \mu\text{g}/\mu\text{gC}$, respectively (Kleindienst et al., 2007), with uncertainty described in
 406 Sect. S2. The fraction of LEVO in WSOC ($0.0994 \mu\text{g}/\mu\text{gC}$) from the BB plume was used for $WSOC_{BB}$ (Ding et
 407 al., 2008). The f_{SOC} value for monoterpenes was scaled up by a factor of 3.1 based on experimental observations,
 408 as these two tracers (HGA+HD-MGA) accounted for 2/9 of the total tracers of monoterpenes, as described in
 409 the supporting information (Kleindienst et al., 2007).

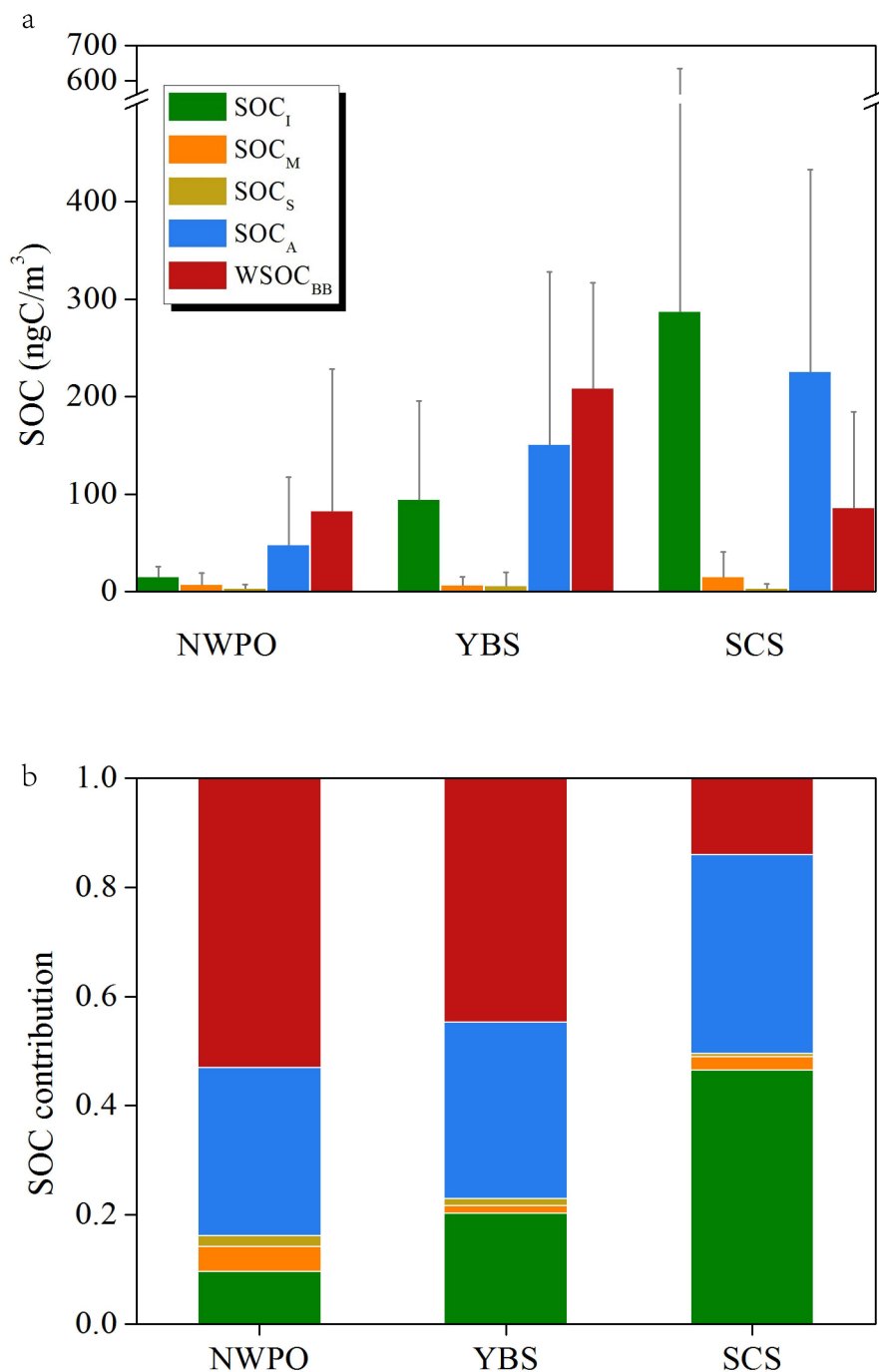
411 Over the SCS, nearly half of the sum of SOC and $WSOC_{BB}$ was in the form of SOC_I (47%), followed by SOC_A
 412 (36%), $WSOC_{BB}$ (14%) and a minor contribution of 2.5% from SOC_M (Fig. 5). This composition pattern over
 413 the SCS could be attributed to abundant biogenic SOA formation in low-latitude tropical marine atmospheres.
 414 Over tropical marine regions, atmospheric oxidation products can account for 47–59% of the total organic
 415 content estimated, with biomass burning emissions making up only 2–7% based on source apportionment using
 416 organic tracers (Fu et al., 2011). A model study by Fu et al. (2012) showed that secondary formation accounts
 417 for as much as 62% of OC estimated using tracers in eastern China in summer. A reverse pattern was observed
 418 over the YBS, with $WSOC_{BB}$ as the dominant contributor (45%) to the sum of SOC and $WSOC_{BB}$, followed by
 419 SOC_A (32%) and SOC_I (20%). The contribution of SOC_M was also minor, at 1.5%. Notably, the chemical
 420 composition observed over the NWPO was similar to that over the YBS, with $WSOC_{BB}$ contributing up to 53%.
 421 In addition, Kang et al. (2018) used the PMF method to identify various sources of OC in marine aerosols over
 422 the ECS such as secondary nitrate, BSOA, BB, and fungal spores.

423 Geographically, the estimated SOC values from BVOCs ranked at the highest level of $306 \pm 343 \text{ ngC}/\text{m}^3$ over the
 424 SCS, decreasing to $107 \pm 99 \text{ ngC}/\text{m}^3$ over the YBS and $24 \pm 22 \text{ ngC}/\text{m}^3$ over the NWPO. The estimates of
 425 aromatic SOC exhibited the same geographic trend, with values of $225 \pm 208 \text{ ngC}/\text{m}^3$ over the SCS, 151 ± 177
 426 ngC/m^3 over the YBS and $48 \pm 69 \text{ ngC}/\text{m}^3$ over the NWPO. Recent modeling results have also shown that
 427 aromatic emissions are the predominant precursors of SOA during springtime in China in comparison with
 428 BVOCs and other AVOCs (Han et al., 2016). Among estimates of $WSOC_{BB}$, the highest values of 209 ± 108
 429 ngC/m^3 were recorded over the YBS, followed by comparable levels of $86 \pm 98 \text{ ngC}/\text{m}^3$ (SCS) and 83 ± 145
 430 ngC/m^3 (NWPO).

431 In our study, the calculated $WSOC_{BB}$ estimate accounted for $4.1 \pm 5.0\%$ and $3.3 \pm 1.7\%$ of measured OC over the
 432 NWPO and YBS, respectively, and these values are higher than that obtained over the ECS during summer
 433 (1.4%) (Kang et al., 2018). Estimated SOC from BVOCs accounted for only $1.5 \pm 1.4\%$ and $1.8 \pm 1.7\%$ to the
 434 measured OC over the NWPO and YBS, respectively, which is lower than that over ECS (4.21%) (Kang et al.,
 435 2018). However, the mean values obtained in this study were similar to the total SOC level estimated using
 436 tracers as a proportion of measured WSOC (4%) during a cruise on the North Pacific and Arctic Oceans,
 437 supposed that WSOC accounted for half of the total OC in atmospheric particles (Ding et al., 2013).

438 The calculated SOC level derived from organic tracers accounted for less than 8% of total measured OC in these
 439 study areas. However, ~~this-these~~ SOC compounds are expected to derive mainly from photochemical reactions
 440 in the gas phase, followed by gas-aerosol partitioning. These compounds likely play an important role in the
 441 growth of newly formed particles alongside pre-existing nucleation mode or Aitken mode particles. However,
 442 most organic matter detected in bulk samples may originate from primary sources, ~~heterogeneousheterogenous~~

443 reactions and in-cloud processing (Ervens et al., 2011; Kanakidou et al., 2005; Nichols, 2016), and these
 444 compounds may be major drivers of the direct climate effects of aerosols, rather than indirect climate effects. In
 445 the future, a comprehensive combination measurement of organic tracers and organic matter with an aerosol
 446 mass spectrometer should be used to elucidate the formation and growth processes of atmospheric nanoparticles.
 447



448
 449 **Figure 5. Average SOC levels calculated using the tracer-SOC/WSOC method over three marine regions**
 450 **(ECS-YBS and NWPO in 2014, SCS in 2017) and their contributions based on five organic tracers**
 451 **measured in this study.**

452 4. Conclusions

453 This study investigated the geographical distributions of tracer-based organic matter observations in TSP
454 collected over two marginal seas of China and the NWPO in the spring season, when the East Asian monsoon
455 carries biogenic and anthropogenic aerosols over these oceanic zones. We found ~~that~~ a significantly large
456 difference in LEVO over the NWPO between two categories of air masses originating from upwind continents
457 or oceanic regions, as Category 1 (continental) contained 13 ± 18 ng/m³ and Category 2 (oceanic) had 2.0 ± 1.8
458 ng/m³; the concentrations of LEVO in Category 2 were closer to the low values reported in the literature. This
459 further implied a large increase in continent-derived BB aerosols in marine atmospheres over the NWPO in
460 recent decades, compared to previous studies. An important question is thereby raised, i.e., does a large increase
461 in continent-derived BB aerosols in marine atmospheres over the NWPO occur continuously and largely in
462 recent decades? Combining the L/M ratios of 19 ± 4 over the NWPO with the calculated air mass back
463 trajectories indicates that the increase was very likely associated with enhanced emissions of BB aerosols from
464 wildfires in Siberia and northeastern China. Moreover, the mean level of BB aerosols over the SCS nearly
465 matched that over the NWPO. The contents of LEVO in Category 2 air masses, derived from oceanic zones over
466 the SCS, were comparable with those reported in the literature, but the mean value was only about a quarter of
467 that in Category 1, representing air masses from upwind continents. However, the limited data available over the
468 SCS in the literature cannot support inferences about whether BB aerosols emitted from upwind tropical forests
469 have increased in recent decades.

470 The concentrations of SOA_I over the SCS were approximately one order of magnitude greater than those
471 observed over the NWPO and several times larger than those over the YBS. The larger values observed over the
472 SCS in Category 1 than in Category 2 were likely driven by high emissions of isoprene from upwind tropical
473 forests and strong solar radiation. The MTLs dominance of SOA_I over the SCS strongly suggested that SOC
474 from BVOCs was generated primarily under low-NO_x conditions. On the other hand, 2-MGA dominance over
475 the YBS implied that most SOC was generated under high-NO_x conditions. Elevated ratios of 2-MGA/MTLs
476 of >1.5 were obtained for 11 of 19 total samples collected over the NWPO, consistent with those reported in the
477 literature. Larger ratios may be attributed to possible emissions of BVOCs in the presence of BB. However, the
478 comparable concentrations of SOA_I in Category 1 and Category 2 samples collected over the NWPO implied a
479 large contribution of SOA_I from marine sources. The aromatic SOA tracers' levels were highest over the SCS,
480 followed by values obtained over the YBS and NWPO. The high values observed over the SCS may be related
481 to strong solar radiation, but the sources of precursors remain unexplained. Based on the concentrations in
482 Category 1 and 2 air samples collected over the SCS and NWPO, mixed sources of aromatic VOCs should exist,
483 including continent-derived precursors, oil exploration and heavy marine traffic.

484 Over the NWPO and the YBS, the estimated WSOC_{BB} levels were nearly equal to the sum of SOC estimated
485 from the oxidation of aromatics and BVOCs. Over the SCS, SOC estimated from the oxidation of BVOCs was
486 significantly larger than the estimated WSOC_{BB}. The geographical difference may be related to emissions of
487 primary particulate organics and gaseous precursors as well as formation processing of secondary organics in
488 various atmospheres.

489 The atmospheric composition of SOA in different geographical locations is, however, highly complex and is
490 regulated by many factors including local meteorological conditions, anthropogenic emissions, plant species,
491 vegetation cover and regional chemistry, and therefore warrants further quantification and analyses. Particularly,
492 whether BB aerosols and other biogenic organic aerosols in marine atmospheres will continuously increase
493 under warming conditions.

494 **Table 1. Sum of organic tracer contents (ng/m³) at different locations worldwide.**

Site	Date	Sampler	LEVO	SOA _I	SOA _M	SOA _S	SOA _A	Reference
Wakayama, Japan (Forest)	August 20–30, 2010, Day	TSP	2.5±2.1	281±274	54.6±50.2	1.2±1.2		(Zhu et al., 2016a)
	Night		1.1±0.9	199±207	36.3±33.6	0.9±0.8		
Across China	summer 2012	Anderson sampler		123±79	10.5±6.6	5.0±4.0	2.9±1.5	(Ding et al., 2014)
Beijing (PKU) (urban site)	summer 2007	PM2.5	37-148	59±32	30±14	2.7±1.0		(Yang et al., 2016)
Beijing (YUFA) (suburban site)			34-149	75±43	32±14	3.9±1.5		
Shanghai (BS) (Suburban site)	Apr-May 2010	PM2.5	88.8±57.2	3.8±3.9	6.1±3.7	1.0±0.7	1.1±0.7	(Feng et al., 2013)
Shanghai (XJH) (Urban site)			58.3±27.5	2.5±1.7	2.7±1.3	0.4±0.3	0.6±0.4	
Mt. Tai	summer 2014	PM2.5		56.4±45.6	34.4±28.4			(Zhu et al., 2017)
Central Pearl River Delta	fall-winter 2007	PM2.5		30.8±15.9	6.6±4.4	0.5±0.6		(Ding et al., 2011)
Central Tibetan Plateau	2012-2013	Anderson sampler		26.6±44.2	1.0±0.6	0.09±0.1	0.3±0.2	(Shen et al., 2015)
Mumbai, India	winter 2007	PM10		4.1±2.4	29±22		0.6±0.6	(Fu et al., 2016)
	summer 2007		1.1±0.7	9.4±4.7	0.05±0.1			
Alaska	Spring 2009	TSP		2.4	3.6	0.9		(Haque et al., 2016)
	2008-2009	TSP		4.1	2.0	1.5		
SYS	Spring 2017	TSP	9.6±8.6	45±54	3.5±6.0	0.07±0.1	1.8±1.7	This study
YBS	Spring 2014	TSP	21±11	15±16	1.6±2.0	0.1±0.3	1.1±1.4	This study
NWPO	Spring 2014	TSP	8.2±14	2.3±1.6	1.6±2.7	0.05±0.09	0.3±0.5	This study
East China Sea	18 May to 12 June 2014	TSP	0.09–64.3 (7.3)	0.15–64.0 (8.4)	0.26–87.2 (11.6)	0.16–17.2 (2.9)		(Kang et al., 2018)
Arctic to Antarctic	July to September 2008; November 2009 to April 2010	TSP	5.4±6.2	8.5±11	3.0±5.0			(Hu et al., 2013a; Hu et al., 2013b)
North Pacific	2003	TSP		0.5±0.4	0.6±0.4	0.06±0.05	0.002±0.005	(Ding et al., 2013)

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496 **Data availability.** Most of the data are shown in supplement. Other data are available by contacting the
497 corresponding author.

498 **Supplement.** The supplement related to this article is available.

499 **Author contributions.** XY, TG and JF conceived and led the studies. TG, JW and JF carried out the
500 experiments and analyzed the data. TG and JF interpreted the results. ZG, JF, HG discussed the results and
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