Response to Anonymous Referee #2

The manuscript reports the spatiotemporal distributions of organic tracers in TSP collected in two marginal seas of China and the NWPO in the spring season and how the East Asian monsoon carries biogenic and anthropogenic aerosols over these oceanic zones. In addition, the authors discussed the origins of SOA_I over the SCS and NWPO. Overall, it is an interesting and inspiring work. However, the follow comments need to be addressed before it can be accepted for publication on ACP.

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

Major comments:

1) Line 152-154: it's better to see if the levo/TSP ratio had been increased. Otherwise it's inconclusive to say the contribution of BB aerosols to particle loading over the NWPO may have increased...

Response：The origin sentence is misleading and has been revised "Using these previous observations as a reference (Table 1), our observations suggested that the BB aerosols from the long-range transport over the NWPO in 2014 largely increased. Thus, an important question is raised, i.e., does the increase occur continuously and largely over the last decades in marine atmospheres over the NWPO?". The LEVO/TSP was 0.02% ± 0.03% (average ± standard deviation) and 0.02% ± 0.01% over the NWPO and over the YBS. It is meaningless to say the contribution of BB aerosols to particle loading over the NWPO.

2) Line 227: the relative contribution of SOA tracers to TSP in category 2 is much larger than that in category 1. Based on the authors' reasoning, is it realistic to infer that marine sources can contribute around 10% of TSP?

Response：We carefully check through the whole manuscript. We are sorry for the misleading, but we cannot find where cause this. In revision, we added "The average contribution of SOA tracers to TSP over the SYS was higher in category 1 (0.4% ± 0.6%) than in category 2 (0.06% ± 0.07%)." And "The average contribution of SOA tracers to TSP over the NWPO was higher in category 1 (0.008% ± 0.005%) than that in category 2 (0.005% ± 0.005%)."

3) Line 294: what are the possible major precursors for DHOPA other than BB emission?

Response: The sentence has been revised as "leaving emissions other than BB emissions, e.g., solvent use, oil exploration, marine traffic, etc., as the major precursors for DHOPA in these marine atmospheres".

4) Line 362: it might be attributable to the different stability of 2-MGA and LEVO?

Response: Thanks for the suggestion. In revision, we added "The decomposition of LEVO reported in literature (Hennigan et al., 2010; Hoffmann et al., 2010; Fraser and Lakshmanan, 2000) may lower the correlation between them. However, whether 2-MGA can decompose in ambient air remains poorly understood."

Reference:

Fraser, M. P. and Lakshmanan, K.: Using levoglucosan as a molecular marker for the long-range transport of biomass combustion aerosols, Environ. Sci. Technol., 34, 4560-4564, https://doi.org/10.1021/es991229l, 2000.

Hennigan, C. J., Sullivan, A. P., Collett Jr., J. L. and Robinson, A. L.: Levoglucosan stability in biomass burning particles exposed to hydroxyl radicals, Geophys. Res. Lett., 37, L09806, https://doi.org/10.1029/2010GL043088, 2010.

Hoffmann, D., Tilgner, A., Iinuma, Y. and Herrmann, H.: Atmospheric stability of levoglucosan: A detailed laboratory and modeling study, Environ. Sci. Technol., 44, 694-699, https://doi.org/10.1021/es902476f, 2010.

Minor comments:

1)Line 28: change "discuss" to "discussed"

2) Line 237: there is a redundant "burning"

3) Line 181: change "surprised" to "surprising"

4) Line 350: better to change "regarding" to "given that"

Response: Done. Thanks for your advice.

Response to Anonymous Referee #3

The manuscript of Guo et al. focuses on quantification of primary and secondary organic tracers in total suspended particles over the Yellow and Bohai seas (YBS) and the South China Sea (SCS) collected both in 2014, and the northwest Pacific Ocean (NWPO) collected in 2017. The authors focused on the long-range transport impact to the marine atmosphere in spring during East Asian monsoon season. Thus, the influence of continental BB aerosols in

72 *marine atmosphere was studied and the contributions of different precursors to the observed*

73 *SOA were quantified using specific tracers. Also, SOA formation pathways in marine*

74 *atmosphere have been discussed and related to the literature data. It is high quality work*

75 *focusing on organic aerosols from remote marine areas which are rare compared with*

76 *continental ones.*

77

78 **Response：** We thank the reviewer's comments and revise our manuscript accordingly.

79

80 *Major comment: Major comment is related to the fact that this study presents a snapshot*

81 *situation at particular marine areas and it is hard to distinguish how representative they are*

82 *for general conclusions. With that in mind, the discussion and conclusions related to the SOA*

83 *change in recent decades are questionable. This is especially addressed to the*

84 *continent-derived BB aerosols affected by accidental intensive open-fire events and or*

85 *specific meteorological conditions. Long-term study is needed to enable conclusions on*

86 *increase/decrease in contribution of continental-derived BB aerosols to remote marine areas*

87 *(L17-19; L152-154; L434-443). Despite the authors did great effort to support their*

88 *discussion with the relevant literature, conclusions arising from there should be done more*

89 *carefully since the authors compare a snapshot situation. Authors should comment on that*

90 *and/or all general conclusions related to above should be avoided.*

91

92 **Response:** We agree with the comments and soften the arguments accordingly. In revision,

93 they have been revised as "The comparison of levoglucosan observed in this study with

94 values from the literature showed that the concentrations of biomass burning aerosols over

95 the NWPO increased largely in 2014. More observations together with the snapshot

96 measurement, however, need to confirm whether the large increase occurred continuously

97 through the last decades."

98

99 "Using these previous observations as a reference (Table 1), our observations suggested that

100 the BB aerosols from the long-range transport over the NWPO in 2014 largely increased.

101 Thus, an important question is raised, i.e., does the increase occur continuously and largely

102 over the last decades in marine atmospheres over the NWPO?"

103

104 "This further implied a large increase in continent-derived BB aerosols in marine

105 atmospheres over the NWPO recently, compared to previous studies. An important question

106 is thereby raised, i.e., does a large increase in continent-derived BB aerosols in marine

107 atmospheres over the NWPO occur continuously and largely in recent decades?"

108

Minor comments

Line 153 Add . . .as a reference (Table 1),

L161-L163 Remove –samples- from the sentence or rewrite

L181 Change –surprised- with to surprise or surprising

Response: Done. Thanks.

L205 I suggest to rewrite the first sentence in figure caption, it is unclearly written

Response:

The caption was rewritten into "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 Management System (FIRMS, https://firms.modaps.eosdis.nasa.gov/). And the base map was from Resource and Environment Data Cloud 210 Platform, DOI: 10.12078/2018110201.".

L212 Replace NWPO with SCS.

Response: Done. Thanks.

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 implied that the contribution from biomass burning emissions to marine aerosols over the NWPO may have increased largely over the last decades.~~ 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 snapshot measurement, however, need to confirm whether the large increase occurred continuously through the last decades. The increase led to the mean value of levoglucosan (8.2 ± 14 ng m^{-3}) observed over the NWPO closer to that over the SCS (9.6 ± 8.6 ng/m^3) and almost half of that over the YBS (21 ± 11 ng/m^3). 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/m^3) several times and approximately one order of magnitude greater than that over the YBS (15 ± 16 ng/m^3) and the NWPO (2.3 ± 1.6 ng/m^3), 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/m^3, 1.1 ± 1.4 ng/m^3 and 0.3 ± 0.5 ng/m^3 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 long-range transport in the atmosphere, which can carry them from the continents to the oceans (Ding et al., 2013; Fu et al., 2011; Kanakidou et al., 2005). For example, BB aerosols from boreal forest wildfires in Russia and China reportedly made an appreciable contribution to atmospheric particle loads observed over the Arctic Ocean and northwestern Pacific Ocean (NWPO) based on specific tracers of BB (Ding et al., 2013). Although open wildfires from forests occur sporadically in terms of strength and occurrence frequency, global warming could be conducive to vegetation fires (Running, 2006) and thus increase emissions of BB aerosols. In this century, nine years were among the ten hottest global years on record, with 2014–2018 being ranked as the top five hottest years (https://www.climatecentral.org/gallery/graphics/the-10-hottest-global-years-on-record). The question is automatically raised: how do BB aerosols in the marine atmosphere in the hottest global years change against those observations previously reported?

In addition to BB aerosols, secondary oxidation of biogenic volatile organic compounds (BVOCs) and anthropogenic VOCs (AVOCs) also contribute to the particulate carbonaceous components of marine atmospheres (Kanakidou et al., 2005). Many field and modeling studies have proposed that secondary organic aerosols (SOAs) arising from the oxidation of phytoplankton-derived isoprene may affect the chemical composition of marine atmospheric aerosols and consequently impact CCN loading and cloud droplet number concentrations (Ekström et al., 2009; Meskhidze and Nenes, 2006; Claeys et al., 2004). Several modeling studies have shown that the NWPO may experience the greatest increases in sea surface temperature and CO_2 input under a future warming climate in the future (John et al., 2015; Lauvset et al., 2017). The Kuroshio Extension current system leads the NWPO to be an active subtropical cyclone basin, promoting biogenic activities (Hu et al., 2018). From the perspective of global change, it is a long-term need to study the dynamic changes in atmospheric aerosols derived from marine sources over the NWPO and adjacent marginal seas of China, as well as their potential effects on climate.

Not limited by phytoplankton-derived isoprene, BVOCs emitted from continental ecosystems and their oxidation products can also affect the atmosphere in remote marine areas through long-range transport (Hu et al., 2013a; Ding et al., 2013; Kang et al., 2018; Fu et al., 2011; Kawamura et al., 2017). BVOCs consist primarily of isoprene, monoterpenes, sesquiterpenes, and their oxygenated hydrocarbons such as alcohols, aldehydes, and ketones (Guenther et al., 2006; Ehn et al., 2014) and account for the majority of the global VOC inventory (Zhu et al., 2016a, b; Heald et al., 2008). However, emissions fluxes and oxidation processes of BVOCs show great variation, depending on global warming and other factors such as regional landscape, other pollutants in the ambient air etc. (Ait-Helal et al., 2014; Hu and Yu, 2013; Peñuelas and Staudt, 2010). Unlike a potential increase in BVOC-derived organics aerosols in marine atmospheres under global warming, anthropogenic VOCs and carbonaceous particles over the continents have been decreased because of effective mitigation of air pollutants in the last decades (Li et al., 2019, Sharma, 2004; Murphy et al., 2011; Zhang et al., 2012). In the northern hemisphere, marine atmospheres are also usually affected by anthropogenic pollutants to some extent, most of which are derived from long-range transport from continents (Kang et al., 2019; Bao et al., 2018; Zhang et al., 2017). The revere trends in BVOC and anthropogenic VOC would change the composition, sources of carbonaceous particles in marine atmospheres. Update observations are thereby needed to reveal the change and service the future study of the impacts.

In this study, we analyzed the concentrations of some typical organic tracers in aerosol samples obtained from three cruise campaigns from the marginal seas of China, including in the South China Sea (SCS) in 2017, Yellow Sea and Bohai Sea (YBS), to the NWPO in 2014, both in springtime. We investigated the influences of BB aerosols from continents over three marine atmospheres, quantified the contributions of various precursors

83 to the observed SOA in marine atmospheres using organic tracers established in the literature, and explored the

84 formation pathways of SOA from their precursors during long-range transport in these hottest global years.

85 Particularly, we conducted a comprehensive comparison of this observation with those reported in literature in

86 terms of long-term variations and geographic distributions of these tracers, etc.

87 **2 Materials and Methods**

88 Total suspended particulate (TSP) samples were collected over the NWPO from 19 March to 21 April 2014,

89 over the YBS from 30 April to 17 May 2014, and over the SCS from 29 March to 4 May 2017. All samples were

90 collected on the upper deck of the R/V Dong Fang Hong II, which sits ~8 m above the sea surface. To avoid

91 contamination from the ship's exhaust, samples were collected only when the ship was sailing, and the wind

92 direction ranged from –90° to 90° relative to the bow. TSP samples were collected on quartz fiber filters

93 (Whatman QM-A) that had been pre-baked for 4 h at 500°C prior to sampling using a high-volume sampler

94 (KC-1000, Qingdao Laoshan Electric Inc., China). The sampling duration was 15–20 h at a flow rate of ~1000 L

95 /min. After sampling, the sample filters were wrapped in baked aluminum foil and sealed in polyethylene bags,

96 then stored at -20°C and transported to the laboratory. Field blanks were collected during each sampling period.

97 However, one sampler was out of service during the cruise on the SCS. As a compromise, cellulose filters

98 (Whatman 41) previously intended for elemental analyses were used for analyses of the organic tracers in TSP.

99 The method for determining the concentrations of tracers was adapted from Kleindienst et al. (2007) and Feng et

100 al. (2013). Briefly, 20 mL dichloromethane/methanol (1:1, v/v) was used for ultrasonic extraction of 40 cm^2 of

101 each filter at room temperature three times. The combined extracts were filtered, dried under a gentle stream of

102 ultrapure nitrogen, and then derivatized with 100 μL N,O-bis-(trimethylsilyl)-trifluoroacetamide (BSTFA,

103 containing 1% trimethylchlorosilane as a catalyst) and 20 μL pyridine at 75°C for 45 min. Gas chromatography

104 mass spectrometry (GC-MS) analyses were conducted with an Agilent 6890 GC/5975 MSD. Prior to solvent

105 extraction, methyl-β-D-xylanopyranoside (MXP) was spiked into the samples as an internal/recovery standard.

106 Hexamethylbenzene was added prior to injection as an internal standard to check the recovery of the surrogates.

107 Like those reported by Feng et al. (2013), the primary organic tracers analyzed in this study included

108 levoglucosan (LEVO), mannosan, and galactosan. Four types of secondary organic tracers were used:

109 isoprene-derived secondary organic tracers (SOA$_I$) including 2-methylglyceric acid (2-MGA), C5-alkene triols

110 (cis-2-methyl-1,3,4-trihydroxy-1-butene,   3-methyl-2,3,4-trihydroxy-1-butene   and

111 trans-2-methyl-1,3,4-trihydroxy-1-butene),   and   MTLs   (2-methylthreitol   and   2-methylerythritol);

112 monoterpene-derived   secondary   organic   tracers   (SOA$_M$)   including   3-hydroxyglutaric   acid   (HGA),

113 3-hydroxy-4,4-dimethylglutaric acid (HDMGA), and 3-methyl-1,2,3-butanetricarboxylic acid (MTBCA); the

114 sesquiterpene-derived   secondary   organic   tracer   (SOA$_S$)   β-caryophyllinic   acid;   and   the   aromatic

115 (toluene)-derived secondary organic tracer (SOA$_A$) 2,3-dihydroxy-4-oxopentanoic acid (DHOPA). LEVO was

116 quantified based on authentic standards in this study. While the SOA tracers without available commercial

117 standards were quantified using methyl-β-D-xylanopyranoside (MXP) as a surrogate. To reduce the uncertainty

118 of quantification, relative response factors of the target tracers to MXP were estimated by comparing the area

119 ratio of typical target ions to MXP to that of total ions in selected samples that showed high concentrations of

120 the target tracers (Feng et al., 2013).

121 Field blanks and laboratory blanks (run every 10 samples) were extracted and analyzed in the same manner as

122 the ambient samples. Target compounds were nearly always below the detection limit in field and laboratory

123 blanks. Recoveries of the surrogate (MXP) were in the range of 70–110%. The reported results were corrected

124 for recovery, assuming that the target compounds had the same recovery as the surrogate. Duplicate analyses

125 indicated that the deviation was less than 15%.

126   However, the substitution of cellulose filters (Whatman 41) during the cruise on the SCS led to increased field

127   blank values for some tracers. The tracer concentrations in those samples were, however, over three times higher

128   than the field blank values, except for those of mannosan and galactosan. Data for mannosan and galactosan

129   were thus not available, nor were the total organic carbon concentrations, for samples collected during the cruise

130   on the SCS.

131   The concentrations of organic carbon (OC) and element carbon (EC) in each sample were analyzed with a DRI

132   2001A thermal/optical carbon analyzer (Atmoslytic Inc., Calabasas, CA, USA) using the IMPROVE

133   temperature program (Wang et al., 2015). All filters before and after sampling were weighted at a glovebox

134   under the controlled ambient temperature and relative humidity. Mass concentrations of TSP, however, should

135   be treated as semi-quantitative results by considering analytic errors of quartz fiber filters (Yao et al., 2009).

136   3. Results and Discussion

137   3.1 Spatiotemporal distributions of LEVO

138   Levoglucosan, mannosan, and galactosan produced by the pyrolysis of cellulose and hemicellulose have been

139   widely used as organic tracers of BB aerosols in ambient air (Ding et al., 2013; Fu et al., 2011; Feng et al.,

140   2013). The mean levels of LEVO in TSP collected during the cruises on the NWPO and the SCS were

141   comparable, at 8.2 ng/m^3 and 9.6 ng/m^3, respectively (Figure S1, Table 1). They were almost half of the mean

142   value of 21 ng/m^3 during the cruise on the YBS, where high concentrations of BB aerosols have been observed

143   in continental atmospheres upwind of the YBS mainly from wildfires and the burning of burning crop residue,

144   wildfire, etc. (Yang et al., 2014; Feng et al., 2012; Feng et al., 2013). Unlike the smaller difference among the

145   means values, the concentration of LEVO fluctuated greatly among TSP samples in each oceanic zone, ranging

146   from 0.5 to 65 ng/m^3 over the NWPO, from 1.0 to 30 ng/m^3 over the SCS and from 2.5 to 42 ng/m^3 over the

147   YBS (Fig. S1). High spatiotemporal variation in LEVO in TSP has also been observed in literature, with

148   concentrations of LEVO fluctuating around 0.2–41 ng/m^3 during Arctic to Antarctic cruises from July to

149   September 2008 and from November 2009 to April 2010 (Hu et al., 2013b). Hu et al. (2013b) also reported the

150   highest LEVO concentrations occurring at mid-latitudes (30°–60° N and S) and the lowest at Antarctic and

151   equatorial latitudes over the several months of sampling. This distinctive geographical distribution was not

152   observed in the present study, as there were no significant differences in LEVO in TSP between the SCS and

153   NWPO (P > 0.05).

154   Narrow spatiotemporal variation in LEVO in TSP has been reported during summer sampling over the North

155   Pacific Ocean and the Arctic in 2003, with maximum and mean values as low as 2.1 ng/m^3 and 0.5 ng/m^3,

156   respectively (Ding et al., 2013). A lower mean value of LEVO of 1.0 ng/m^3 has also been reported in the spring

157   over the island of Chichi-jima from 2001 to 2004 (Mochida et al., 2010), while the levels increased to 3.1 ± 3.7

158   ng/ m^3 in TSP collected on the island of Okinawa in 2009–2012 (Zhu et al., 2015). Using these previous

159   observations as a reference, our observations suggested that the contribution of BB aerosols to particle loading

160   over the NWPO may have increased continuously and largely over the last decades. Using these previous

161   observations as a reference (Table 1), our observations suggested that the BB aerosols from the long-range

162   transport over the NWPO in 2014 largely increased. Thus, an important question is raised, i.e., does the increase

163   occur continuously and largely over the last decades in marine atmospheres over the NWPO?

164   Due to the lack of BB sources in oceans, large spatiotemporal variation in the concentrations of LEVO in the

165   marine atmosphere may be related to the long-range transport of atmospheric particles from continents. Thus, 72

166   h back trajectories of air masses at a height of 1000 m during our sampling periods (Figs. 1, 2) were calculated

167 using the HYSPLIT model (https://ready.arl.noaa.gov/HYSPLIT). Based on the calculated back trajectories, TSP

168 samples could be classified into two categories with Category 1 representing continent-derived aerosol samples

169 and Category 2 being ocean-derived aerosol samples. All 12 samples collected over the YBS fell into Category 1

170 (Fig. 2). Half (11/19) of the samples collected over the NWPO were classified into Category 1 (Fig. 1). A

171 significant difference ($p < 0.05$) was obtained between the concentrations of LEVO in Category 1 (13 ± 18 ng/m^3)

172 and Category 2 (2.0 ± 1.8 ng/m^3) ~~samples~~ over the NWPO. The values in Category 2 were closer to the

173 springtime observations reported by Mochida et al. (2010) and Zhu et al. (2015) as well as the summer

174 observations reported by Ding et al. (2013), reflecting the marine background value less affected by continental

175 air masses. On the other hand, the much higher values in Category 1 than Category 2 further indicated a large

176 increase in contribution of BB aerosols being transported from the continents to the remote marine atmosphere

177 in 2014.

178 On 11 April 2014 over the NWPO, an episode of high LEVO concentration of 65 ng/m^3 occurred (Fig. 1). Like

179 LEVO, the concentrations of galactosan and mannosan in the sample were also the highest among all samples

180 collected over the NWPO. This sample was collected in the oceanic zone, approximately 500 km from the

181 continent of Japan. A combination of air mass back traceries and NASA's FIRMS Fire Map indicated strong BB

182 aerosol emissions from intense fire events in Siberia, followed by long-range transport with the westerly wind as

183 the major contributors to this anomaly (Fig. 1). A similar episodic concentration of LEVO of 27 ng/m^3 in TSP

184 was observed once previously over the NWPO during a circumnavigation cruise (Fu et al., 2011). By combining

185 satellite data with other observations, many studies in literature have found that BB aerosols from major forest

186 fires and smoke events in Siberia could be transported downwind to remote marine regions not only in spring,

187 but also in summer (Generoso et al., 2007; Ding et al., 2013; Huang et al., 2009). In a few cases, BB aerosols

188 have been reported to have reached as far as the adjacent Arctic region (Warneke et al., 2010; Generoso et al.,

189 2007). Van der Werf et al. (2006) estimated the emissions of BB aerosols from Eurasia to be much larger than

190 those from North America. Thus, it is not ~~surprised~~ surprising that the concentrations of LEVO over the NWPO

191 were much higher than those over the eastern North Pacific and western North Atlantic at similar latitudes (Hu

192 et al., 2013b).

193 In addition, both galactosan and mannosan showed strong linear correlations with LEVO ($R^2 = 0.98$, $p < 0.05$)

194 in TSP collected over the NWPO and YBS in this study. These strong correlations indicate that the three tracers

195 were probably derived from the same BB sources. Previous studies have reported LEVO/mannosan (L/M) ratios

196 of 3–10, 15–25, and 25–40 from softwood, hardwood, and crop-residue burning, respectively (Kang et al., 2018;

197 Zhu et al., 2015). The calculated L/M ratios in TSP collected over the NWPO were 19 ± 4 in this study, which

198 implies dominant contributions from herbaceous plants and hardwood. The calculated L/M ratios in TSP

199 collected over the YBS were 14 ± 11, indicating mixed sources.

200 In all, 5 of 13 samples collected over the SCS were classified into Category 1, with air masses identified as

201 originating from either the continental areas of South China or the Philippines (Fig. 2). The concentration of

202 LEVO fluctuated around 17 ± 12 ng/m^3 in Category 1 but decreased to 3.6 ± 3.4 ng/m^3 in Category 2. However, no

203 significant differences were found between categories due to the large variation in LEVO concentration among

204 the limited samples in Category 1 ($p > 0.05$). Forest fires occur accidentally, leading to the large variation in

205 LEVO in Category 1. Southern Asia has been reported to be one of the greatest emissions sources of BB

206 aerosols worldwide (van der Werf et al., 2006), which likely led to the higher mean value of LEVO in Category

207 1. However, the LEVO level observed over the SCS in Category 2 was closer to that reported from low-latitude

208 regions (2.7 ± 1.1 ng/m^3, Table 1) collected during a global circumnavigation cruise (Hu et al., 2013b). Hu et al.

209 (2013b) argued that their low observed concentrations may have been associated with intense wet deposition,

210 degradation as well as intensive moist convection that occurred in the tropical region during their summer cruise.

211 Unfortunately, no previous observations of LEVO in spring can allow us analyzing the long-term variation in
212 contribution of BB aerosols therein. However, this observation can be used for future comparison.
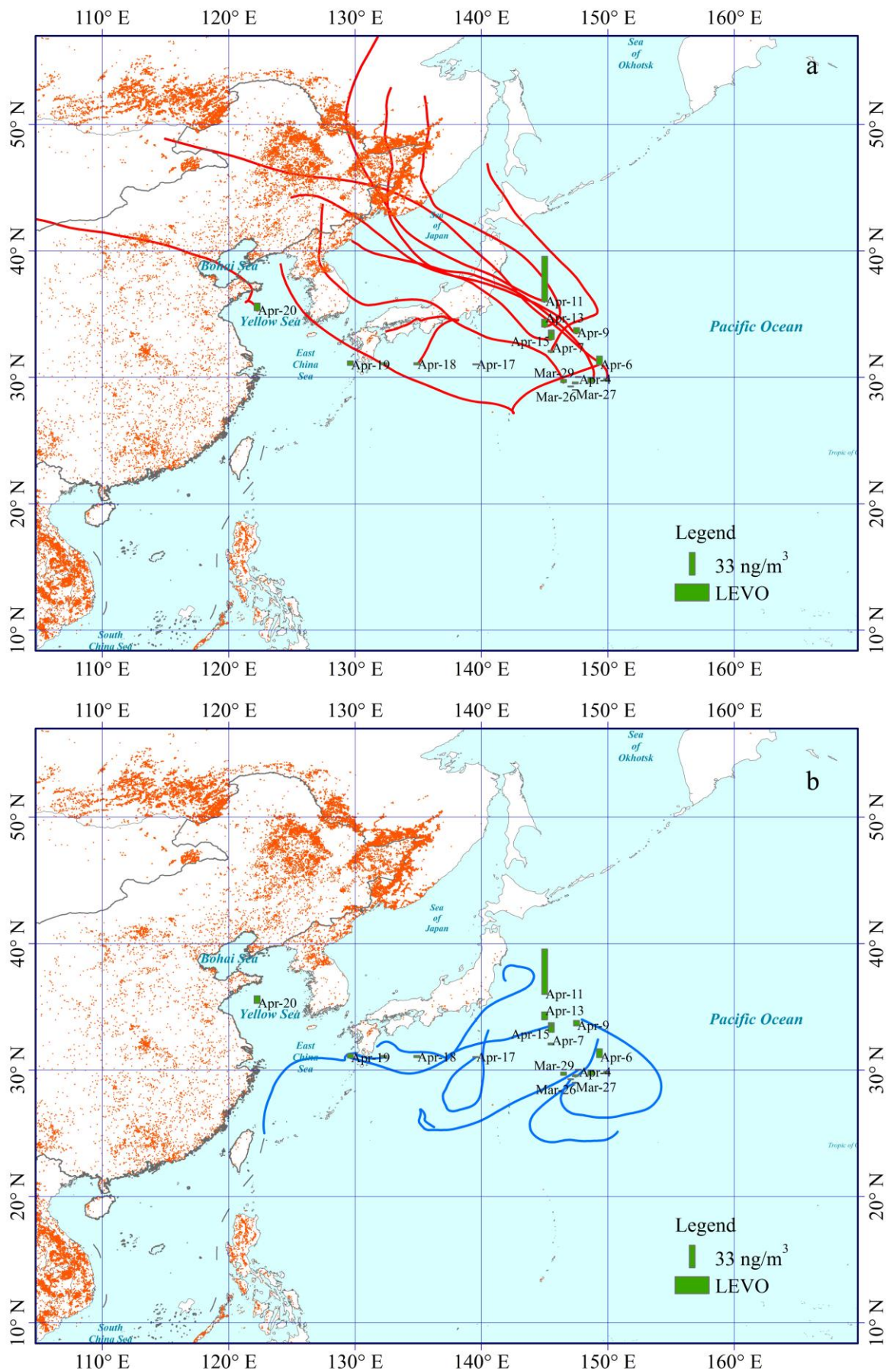
213
214 **Figure 1.** ~~Spatial distribution of LEVO over the NWPO (2014) and BTs corresponding to the samples; the~~
215 ~~samples with the backward trajectories (red lines) indicate land-influenced aerosols (a, Category 1) and~~

216 ~~the blue line denotes ocean-influenced aerosols collected (b, Category 2). The red dots represent the~~
217 ~~locations of fires from Fire Information for Resource Management System (FIRMS,~~
218 ~~https://firms.modaps.eosdis.nasa.gov/). The base map was from Resource and Environment Data Cloud~~
219 ~~Platform, DOI: 10.12078/2018110201.~~ Spatial distribution of LEVO in TSP over the NWPO in spring of
220 2014 and 72-hrs back trajectory associated with each TSP sample. The red lines represent that air masses
221 can be derived from the continent (a, Category 1); the blue lines represent that air masses may be derived
222 mainly from the oceans (b, Category 2). The red dots represent the locations of fires from Fire
223 Information for Resource Management System (FIRMS, https://firms.modaps.eosdis.nasa.gov/). And the
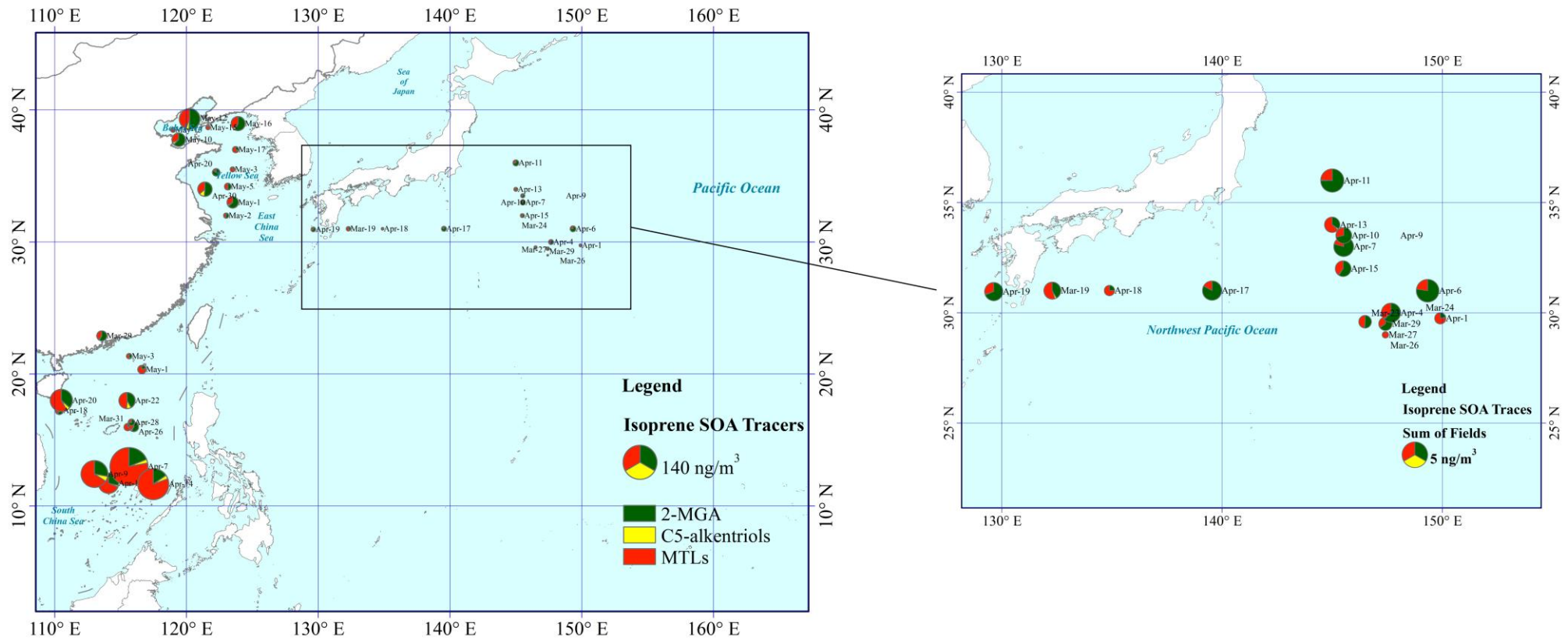224 base map was from Resource and Environment Data Cloud 210 Platform, DOI: 10.12078/2018110201.

225
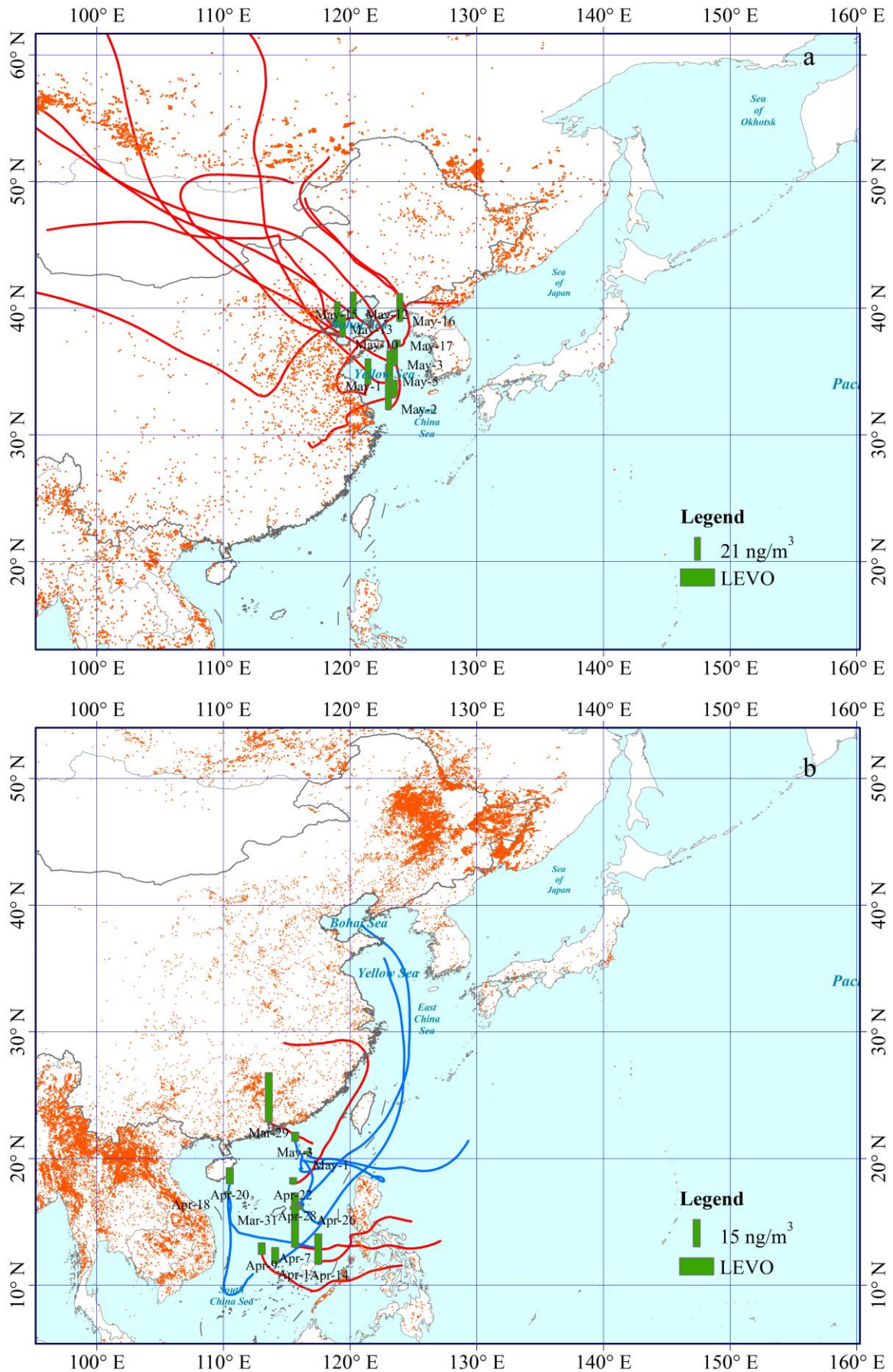
Figure 2. Spatial distribution of LEVO over the YBS (a, 2014), and ~~NWPO~~ SCS (b, 2017), detailed
information descripted in Figure 1. And the base map was from Resource and Environment Data Cloud
210 Platform, DOI: 10.12078/2018110201.

229

3.2 Spatiotemporal distributions of SOA$_I$ tracers

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

When the sum of SOA$_I$ tracers in each sample was examined separately according to the air mass source, a significant difference was found over the SCS between Category 1 (85 ± 66 ng/m^3) and Category 2 (19 ± 22 ng/m^3), with significance at $p < 0.01$. The average contribution of SOA tracers to TSP mass concentration 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 were 2.7 ± 1.8 ng/m^3 in Category 1 and 1.7 ± 1.0 ng/m^3 in Category 2 over the NWPO, where no significant difference between the two categories was found ($p > 0.05$). The average contribution of SOA tracers to TSP mass concentration over the NWPO was higher in category 1 ($0.008\% \pm 0.005\%$) than that in category 2 ($0.005\% \pm 0.005\%$). Supposed that concentrations of the tracers in Category 2 were completely contributed by marine sources, it can be inferred that SOA$_I$ carried by continental air masses increased sharply over the SCS. However, it was not the case over the NWPO. Because all samples over the YBS fell into Category 1, this comparison could not be made for the YBS.

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

254   3.3 Spatiotemporal distributions of SOA$_M$, SOA$_s$ tracers

255   The sum of SOA$_M$ tracers including HGA, HD-MGA, and MBTCA was greatest over the SCS region (3.5±6.0

256   ng/m^3), where the concentration was approximately double that over the YBS (1.6±2.0 ng/m^3) and NWPO

257   regions (1.6±2.7 ng/m^3) (Fig. S1), but no significant differences were identified between any two campaigns.

258   The concentrations of SOA$_M$ tracers were almost one magnitude lower than those of SOA$_I$ tracers. Due to the

259   unique contribution of terpene-derived SOA to nucleation and growth of newly formed particles in the

260   atmospheres (Gordon et al., 2017; Zhu et al., 2019; Ehn et al., 2014), the SOA$_M$ may primarily cause indirect

261   climate effects rather than direct effects of aerosols in the marine atmosphere. The difference in mean SOA$_M$

262   concentration between the SCS and NWPO narrowed to a factor of two, in contrast to the differences of

263   approximately one order of magnitude in mean SOA$_I$ between the two types of atmospheres. The precursors of

264   SOA$_M$ tracers derive mainly from coniferous forests (Duhl et al., 2008) and the decreasing proportion of

265   coniferous forests in subtropical and tropical regions may partially explain the smaller spatial difference in

266   SOA$_M$ tracers over the SCS compared to the YBS and NWPO. However, the comparable SOA$_M$ levels over the

267   YBS and NWPO have not yet been explained.

268   Only three SOA$_M$ tracers were measured in this study, but other SOA$_M$ tracers have been measured and reported

269   in marine atmospheres (Kang et al., 2018; Fu et al., 2011). In order to compare our results with the total amount

270   of SOA$_M$ tracers in the literature, the total amounts measured in this study were multiplied by a factor of 3.1

271   (described in supporting information Sect. S1, Fig. S4) according to the chamber results obtained by Kleindienst

272   et al. (2007). The adjusted values over the SCS were closer to the mean of 11.6 ng/m^3 observed over the ECS

273   (Kang et al., 2018) and the lower values of 9.80–49.0 ng/m^3 observed among 12 continental sites in China (Ding

274   et al., 2016). The adjusted total amounts of SOA$_M$ over the NWPO and YBS were comparable to previous

275   observations of 3.0±5.0 ng/m^3 collected from the Arctic to Antarctic in 2008-2010 (Hu et al., 2013a), but much

276   higher than observations of 63±49 pg/m^3 over the North Pacific and Arctic in 2003 (Ding et al., 2013). This may

277   also imply a substantial increase in SOA$_M$ in the last decades, although more investigations are needed to

278   confirm.

279   β-Caryophyllene is a major sesquiterpene emitted from plants such as Scots pine and European birch (Duhl et al.,

280   2008; Tarvainen et al., 2005). β-Caryophyllinic acid is formed through the ozonolysis or photo-oxidation of

281   β-caryophyllene. The highest levels of β-caryophyllinic acid were observed over the YBS (0.13±0.03 ng/m^3),

282   followed by the SCS (0.08±0.11 ng/m^3) and NWPO (0.05±0.09 ng/m^3) (Fig. S1). The spatial distribution of

283   β-caryophyllinic acid clearly did not follow the general trend of biogenic SOA, with the highest values over the

284   SCS followed by the YBS. Compared to values from the literature, our results are much higher than those over

285   the North Pacific and Arctic Oceans (2.4±5.4 pg/m^3) (Ding et al., 2013) but much lower than observations over

286   the East China Sea reported by Kang et al. (2018), where β-caryophyllinic acid was reported to be in the range

287   of 0.16–17.2 ng/m^3 with a mean of 2.9 ng/m^3. The large differences in β-caryophyllinic acid content observed in

288   various campaigns remains unexplained.

289   3.4 Spatiotemporal distributions of SOA$_A$ tracers

290   When the concentrations of DHOPA in TSP were examined, the highest concentrations occurred over the SCS

291   (1.8±1.7 ng/m^3), followed by the YBS (1.1±1.4 ng/m^3), and the lowest values were recorded in the NWPO

292   region (0.3±0.5 ng/m^3) (Fig. S1). The decreasing extent of the DHOPA decrease from the SCS to the NWPO

293   was approximately three times less than that of SOA$_I$ tracers but approximately three times larger than that of

294 SOA$_M$ tracers. Ding et al. (2017) reported annual averages of DHOPA among various sites in China, which

295 ranged from 1.2 to 8.8 ng/m^3. The concentrations of DHOPA observed over the SCS and the YBS were similar

296 to the lower values observed in upwind continental atmospheres.

297 Formation of DHOPA depends on the molecular structures of aromatics, as well as concentrations of free

298 radicals and oxidants, etc. (Li et al., 2016; Henze et al., 2008). The mean value of DHOPA in Category 1

299 (0.43±0.65 ng/m^3) was nearly twice that in Category 2 (0.20±0.31 ng/m^3) over the NWPO (p > 0.05). With two

300 samples with high DHOPA (1.2, 2.1 ng/m^3) in Category 1 to be excluded, the recalculated average DHOPA

301 decrease down to 0.17±0.21 ng/m^3. The continent-derived DHOPA seemingly yielded a minor contribution to

302 the observed values over the NWPO, except during strong long-range transport episodes. Similarly, the mean

303 values of DHOPA were same in Category 1 (1.8±2.1 ng/m^3) and Category 2 (1.8±1.5 ng/m^3) samples collected

304 over the SCS and no significant differences were observed between two categories. Much stronger UV radiation

305 occurs over the SCS than the YBS, which may contribute to the elevated DHOPA level over the SCS. Aside

306 from continent-derived precursors, oil exploration and heavy marine traffic over the SCS are also potential

307 contributors to the higher DHOPA levels therein, and this link requires further investigation. Previous field

308 observations in China have demonstrated that biofuel or biomass combustion emissions act as important sources

309 of aromatics in the atmosphere (Zhang et al., 2016), as evidenced by the association between the nationwide

310 increase in DHOPA during the cold period and the enhancement of BB emissions (Ding et al., 2017). In this

311 study, no linear correlation was obtained between DHOPA and LEVO in samples collected over the SCS and

312 other two campaigns, leaving emissions other than BB emissions, e.g., solvent use, oil exploration, marine

313 traffic, etc., as the major precursors for DHOPA in these marine atmospheres leaving emissions other than BB

314 emissions as the major precursors for DHOPA in these marine atmospheres (Li et al., 2013).

315 3.5 Causes for high photochemical yields of SOA$_I$ over the SCS

316 Because higher concentrations of SOA$_I$ were observed in TSP samples collected over the SCS, the composition

317 of SOA$_I$ tracers was further investigated in terms of their formation pathways and sources. Based on the results

318 of chamber experiments, Surratt et al. (2010) proposed different formation mechanisms for 2-MGA and MTLs.

319 2-MGA is a C4-dihydroxycarboxylic acid, which forms through a high-NO$_x$ pathway. MTLs and C5-alkene

320 triols are mainly products of the photooxidation of epoxydiols of isoprene under low-NO$_x$ conditions.

321 MTLs acted as the dominant compounds among SOA$_I$ tracers in most TSP samples collected over the SCS, with

322 concentrations of 31±42 ng/m^3 (Fig. 3). The ratio of 2-MGA/MTLs ranged from 0.2 to 3.1, with a median value

323 of 0.6. The ratio exceeded the unity in only 4 of 13 samples. This result allowed us to infer that the observed

324 SOA$_I$ tracers were generated mainly under low-NO$_x$ conditions. Although the concentration of

325 2-methylerythritol was nearly double that of 2-methylthreitol, they were highly correlated (R^2 = 0.99, p < 0.05)

326 because of their shared formation pathway. Satellite data showed that the NO$_2$ levels in South China and the

327 Philippines were low, except in a few hotspots (Fig. S2). Such low-NOx conditions favor the formation of

328 MTLs rather than 2-MGA over the tropical SCS. The isoprene emitted from plants growing on oceanic islands

329 may also undergo chemical conversion to SOA under low-NOx conditions, and low-NOx conditions are always

330 expected in remote marine atmospheres (Davis et al., 2001).

331 In general, zonally and monthly averaged OH concentrations around 15°N are ~50% were greater than those

332 around 35 °N (Bahm and Khalil, 2004). Thus, enhanced formation of MTLs is theoretically expected under the

333 strong UV radiation of tropical regions. However, no significant correlation between the concentrations of

334 MTLs and UV radiation was obtained over the SCS (data not shown) possibly due to the influences of various

335 air masses. A field study showed that MTL yields were positively correlated with ambient temperature in

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

In addition, the MTLs concentration in Category 1 (62 ± 55 ng/m^3) was larger than that ~~for~~ in Category 2 (11 ± 14 ng/m^3). The more abundant MTLs associated with Category 1 was most likely related to long-range transport of these chemicals from upwind continental areas, the oxidation of continental precursors in the marine atmosphere, or both. Large emissions of isoprene were expected from tropical forests upwind of the SCS due to the high vegetation coverage and high ambient temperature of such areas (Ding et al., 2011; Rinne et al., 2002). Global estimates show tropical trees to be responsible for ~80% of terpenoid emissions and ~50% of other VOC emissions (Guenther et al., 2012).

In a clean marine atmosphere, phytoplankton is the sole source of isoprene emissions over the oceans (Bonsang et al., 1992; Broadgate et al., 1997). Chlorophyll-a has been widely employed as a measure of phytoplankton abundance and a proxy for predicting isoprene concentrations in water (Hackenberg et al., 2017). The satellite-derived chlorophyll-a level during the study period over the SCS was below 0.45 mg/m^3, excluding coastal areas (Fig. S3). The MTLs observations of 11 ± 14 ng/m^3 in Category 2 should be considered as the upper limitation value derived from marine phytoplankton in the SCS. Although air masses differed between Categories 1 and 2, a good correlation was obtained between MTLs and 2-MGA when the data in the two categories was pooled for analyses ($R^2 = 0.77$, $P < 0.01$). This strong correlation indicated these tracers are primarily formed through shared pathways. However, this correlation was poor over the NWPO, as discussed below.

3.6 Origin and formation of SOA_I over the NWPO

Over the NWPO, the concentration of 2-MGA was 1.6 ± 1.5 ng/m,^{3} which was generally dominant among SOA_I tracers, followed by MTLs (0.7 ± 0.3 ng/m^3) and C5-alkene triols (0.03 ± 0.02 ng/m^3). When the ratio of 2-MGA/MTLs was further examined, it varied greatly from <0.1 to 6.3, with a median value of 2.1. Most ratios observed over the NWPO in this study were far greater than the values of 0.18–0.59 reported by Hu et al. (2013a) from a global circumnavigation cruise, and also greater than 0.87–1.8 reported in urban areas of California (Lewandowski et al., 2013) and the maximum value of 2.0 obtained over the YBS. Ding et al. (2013) also reported ratios that fluctuated greatly from 0.5 to 10 with a median value of 3.3 during a summer cruise in the NWPO and Arctic Ocean in 2003. The large 2-MGA/MTL ratios over the NWPO appeared to be highly consistent over two independent sampling campaigns.

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

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

378   Merlet, 2001). Ding et al. (2013) thus argued that an increase in emissions of isoprene in the presence of BB,

379   followed by its chemical conversion under high-NO_x conditions, may lead to transport over thousands of

380   kilometers and hold at the detectable concentrations in the remote marine atmosphere over the NWPO. The

381   same argument may hold true for the elevated ratios of 2-MGA/MTLs observed over the NWPO in this study

382   (Fig. 4). However, we did not find a significant correlation between 2-MGA and LEVO over the NWPO. <u>The</u>

383   <u>decomposition of LEVO reported in literature (Hennigan et al., 2010; Hoffmann et al., 2010; Fraser and</u>

384   <u>Lakshmanan, 2000) may lower the correlation between them. However, whether 2-MGA can decompose in</u>

385   <u>ambient air remains poorly understood.</u>

386   On the other hand, the ratios of 2-MGA/MTLs in 3 of 19 samples collected over the NWPO were below 0.5

387   (Figure 4). In these cases, the oxidation of isoprene under low-NOx conditions likely dominated the generation

388   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

389   from the oxidation of isoprene under low-NOx conditions and high-NOx conditions. As the major formation

390   pathways of 2-MGA and MTLs varied greatly among samples, no significant correlation ($R^2 = 0.12$, $p > 0.05$)

391   was obtained between 2-MGA and MTLs over the NWPO. Recall that the tracer values of SOA_I were 2.7±1.8

392   ng/m^3 in Category 1 and 1.7±1.0 ng/m^3 in Category 2. This implied that SOA_I derived from marine sources was

393   comparable to that derived from the continent outflows.
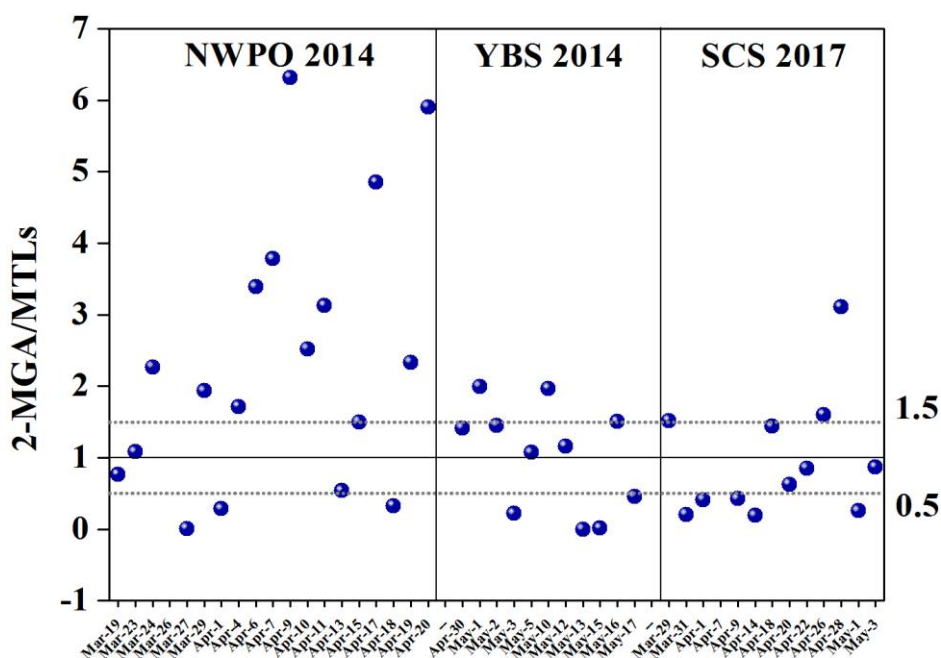
394

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

396   3.7 Source apportionment of secondary organic carbon (SOC)

397   The tracer-based approach developed by Kleindienst et al. (2007) was applied to estimate the concentrations of

398   SOC and $WSOC_{BB}$, as follows:

$$[SOC] = \frac{\sum_i [tri]}{f_{SOC}}$$

399   (1)

$$[WSOC_{BB}] = \frac{C_{tracer}}{f_{tracer/WSOC_{BB}}}$$

(2)

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

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

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

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

The calculated SOC level derived from organic tracers accounted for less than ~~6~~8% of total measured OC in these study areas. However, this SOC compounds are expected to derive mainly from photochemical reactions in the gas phase, followed by gas-aerosol partitioning. These compounds likely play an important role in the growth of newly formed particles alongside pre-existing nucleation mode or Aitken mode particles. However, most organic matter detected in bulk samples may originate from primary sources, heterogonous reactions and

in-cloud processing (Ervens et al., 2011; Kanakidou et al., 2005; Nichols, 2016), and these compounds may be major drivers of the direct climate effects of aerosols, rather than indirect climate effects. In the future, a comprehensive combination measurement of organic tracers and organic matter with an aerosol mass spectrometer should be used to elucidate the formation and growth processes of atmospheric nanoparticles.
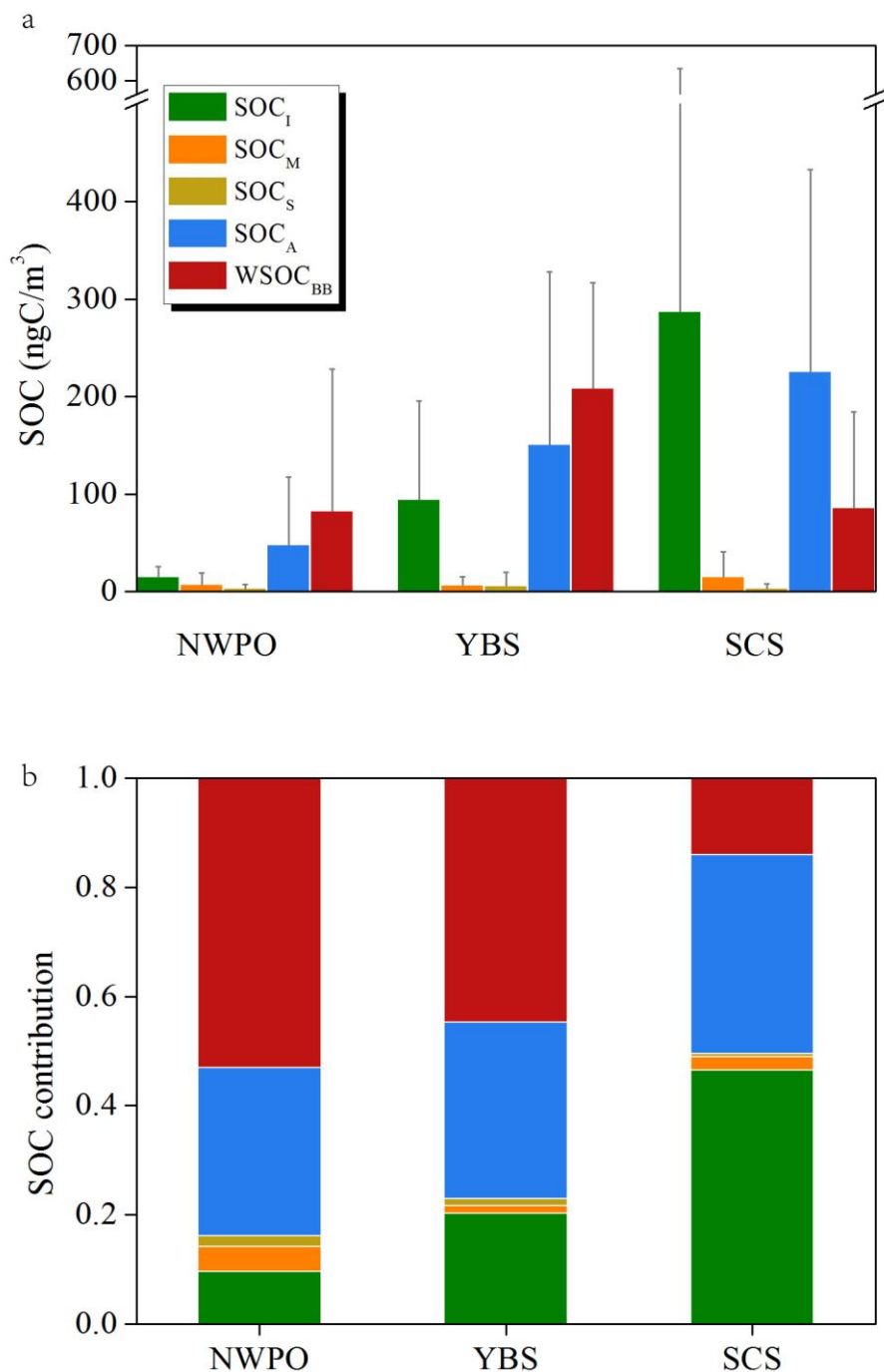
Figure 5. Average SOC levels calculated using the tracer-SOC/WSOC method over three marine regions (ECS and NWPO in 2014, SCS in 2017) and their contributions based on five organic tracers measured in this study.

4. Conclusions

This study investigated the geographical distributions of tracer-based organic matter observations in TSP collected over two marginal seas of China and the NWPO in the spring season, when the East Asian monsoon carries biogenic and anthropogenic aerosols over these oceanic zones. We found that a significantly large difference in LEVO over the NWPO between two categories of air masses originating from upwind continents or oceanic regions, as Category 1 (continental) contained 13 ± 18 ng/m^3 and Category 2 (oceanic) had 2.0 ± 1.8 ng/m^3; the concentrations of LEVO in Category 2 were closer to the low values reported in the literature. This further implied a large increase in ~~contribution of~~ continent-derived BB aerosols ~~to~~ in marine atmospheres over the NWPO in recent decades, compared to previous studies. An important question is thereby raised, i.e., does a large increase in continent-derived BB aerosols in marine atmospheres over the NWPO occur continuously and largely in recent decades? Combining the L/M ratios of 19 ± 4 over the NWPO with the calculated air mass back trajectories indicates that the increase was very likely associated with enhanced emissions of BB aerosols from wildfires in Siberia and northeastern China. Moreover, the mean level of BB aerosols over the SCS nearly matched that over the NWPO. The contents of LEVO in Category 2 air masses, derived from oceanic zones over the SCS, were comparable with those reported in the literature, but the mean value was only about a quarter of that in Category 1, representing air masses from upwind continents. However, the limited data available over the SCS in the literature cannot support inferences about whether BB aerosols emitted from upwind tropical forests have increased in recent decades.

The concentrations of SOA$_I$ over the SCS were approximately one order of magnitude greater than those observed over the NWPO and several times larger than those over the YBS. The larger values observed over the SCS in Category 1 than in Category 2 were likely driven by high emissions of isoprene from upwind tropical forests and strong solar radiation. The MTLs dominance of SOA$_I$ over the SCS strongly suggested that SOC from BVOCs was generated primarily under low-NO$_x$ conditions. On the other hand, 2-MGA dominance over the YBS implied that most SOC was generated under high-NO$_x$ conditions. Elevated ratios of 2-MGA/MTLs of >1.5 were obtained for 11 of 19 total samples collected over the NWPO, consistent with those reported in the literature. Larger ratios may be attributed to possible emissions of BVOCs in the presence of BB. However, the comparable concentrations of SOA$_I$ in Category 1 and Category 2 samples collected over the NWPO implied a large contribution of SOA$_I$ from marine sources. The aromatic SOA tracers' levels were highest over the SCS, followed by values obtained over the YBS and NWPO. The high values observed over the SCS may be related to strong solar radiation, but the sources of precursors remain unexplained. Based on the concentrations in Category 1 and 2 air samples collected over the SCS and NWPO, mixed sources of aromatic VOCs should exist, including continent-derived precursors, oil exploration and heavy marine traffic.

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

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

493 **Table 1. Sum of organic tracer contents (ng/m^3) 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 Ocean and the Arctic	2003	TSP	0.5±0.4	0.6±0.4	0.06±0.05	0.002±0.005	494 (Ding et al., 2013)

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

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

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