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Key Points:
- Long-term observations of BB tracers over the western North Pacific
- BB tracer concentrations are changing according to atmospheric circulation
- The period of the high concentrations was prolonged from 2006 to 2013

Supporting Information:
- Readme
- Figure S1a
- Figure S1b
- Figure S2a
- Figure S2b
- Figure S2c
- Figure S2d
- Figures S3a–S3d

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Thirteen years of observations on biomass burning organic tracers over Chichijima Island in the western North Pacific: An outflow region of Asian aerosols

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Abstract East Asia is the world's greatest source region for the emission of anthropogenic aerosols and their precursors due to the rapid industrialization and intensive biomass burning (BB) activities. BB emits specific organic tracers such as levoglucosan, mannosan, and galactosan, which are produced by pyrolysis of cellulose and hemicellulose and then transported downwind to the western North Pacific by westerly winds. Here we present long-term observations of BB tracers over the remote Chichijima Island in the western North Pacific (WNP) from 2001 to 2013. Elevated concentrations of BB tracers by an order of magnitude were found in midautumn to midspring with winter maxima, which are strongly involved with the atmospheric transport by westerly winds from the Asian continent to the WNP, as supported by backward trajectory analyses. Throughout the observations, we found an increase in the averaged concentrations of BB tracers from 2006 to 2013, which is mainly caused by enhanced BB events in Asian urban and rural areas, as supported by enhanced fire/hot spots in East Asia via satellite images. We also found that the period of the high concentrations was prolonged from 2006 to 2013. Comparison between monthly averaged concentrations of BB tracers and backward air mass trajectories clearly demonstrates that the winter/spring maxima over Chichijima are involved with the seasonal shifting of atmospheric circulation followed by downwind transport of BB aerosols to the WNP. High abundances of BB tracers over the WNP indicate that BB-laden air masses can be transported to remote marine environments.

1. Introduction

Atmospheric transport is the most important delivery pathway for various chemical species from land to open ocean [*Simoneit*, 2002, 2004]. Specific organic tracers can provide useful information on the sources of atmospheric particles [*Fraser and Lakshmanan*, 2000; *Fu et al.*, 2012; *Leithead et al.*, 2006]. Biomass burning (BB) significantly contributes to various organic compounds in atmospheric aerosols [*Crutzen and Andreae*, 1990; *Mayol-Bracero et al.*, 2002; *Mazzoleni et al.*, 2007; *Simoneit et al.*, 2004a; *Sullivan et al.*, 2008]. Levoglucosan, mannosan, and galactosan are specific organic tracers that are produced by pyrolysis of cellulose and hemicellulose [*Simoneit*, 2002]. They are emitted to the air from BB as aerosol particles and then subjected to long-range atmospheric transport downwind of the source regions [*Conte and Weber*, 2002; *Engling et al.*, 2013; *Simoneit and Elias*, 2000]. Long-range transport is a key process for global distribution of organic aerosols from the continent to remote regions. The specific BB tracers provide a useful tool for the assessment of BB particles over the remote ocean [*Simoneit et al.*, 1999].

In East Asia, forest fires, domestic BB for cooking and space heating, and field burning of agricultural residues occur throughout the year, whereas biomass burning in South America and Africa occurs more frequently during dry season [e.g., *Heald et al.*, 2003]. Globally, BB emits significant amounts of gases and particles into the atmosphere (estimated to be 9870 Tg carbon dioxide, 279 Tg carbon monoxide, 107 Tg methane, 2.54 Tg black carbon, and 10.4 Tg organic carbon in the year 2000) [*Streets et al.*, 2003a]. The growing emissions of industrial pollutants, BB products, fossil fuel combustion products, and black carbon are also occurring due to increased economic growth in East Asia [*Lin et al.*, 2014]. Continuously rapid industrialization in East Asia enhances the emission of pollutants that are transported across the North Pacific to North America [*Bailey et al.*, 2000; *Bey et al.*, 2001; *Jaffe et al.*, 1999, 2003]. The transpacific

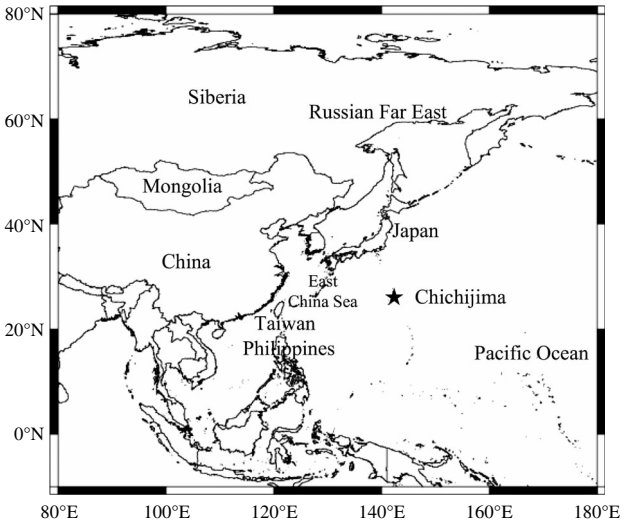


Figure 1. Locality map of Chichijima Island (27°04′N, 142°13′E) with the WNP and Asian continent.

transport of air pollutants has been reported in previous studies [*Jacob et al.*, 2003; *Jaffe et al.*, 2004; *Wang et al.*, 2006; *Warneke et al.*, 2009; *Wilkening*, 2001].

Chichijima Island is located in the western North Pacific (WNP) on the outflow region of Asian dust and pollutants [*Simoneit et al.*, 2004b; *Wang et al.*, 2009]. This island is located in the boundary of westerly and trade wind regimes. From midautumn to midspring, westerly winds dominate over the WNP, delivering natural and anthropogenic aerosols from the Asian continent to the Pacific Ocean, passing over the Russian Far East, East and Northeast Asia, the Sea of Japan, and the main island of Japan [*Kawamura et al.*, 2003; *Mochida et al.*, 2003a, 2003b]. In contrast, during midspring to midautumn, the high ambient temperature over Siberia develops a low-pressure system that drives the transport of warm and moist marine air masses from the central Pacific to the Asian continent. Due to the low population and small area of Chichijima, the local BB emissions are insignificant [*Chen et al.*, 2013]. Therefore, the geographical situation of Chichijima is ideal for studying the BB aerosol loading over the WNP and its impact on regional to global climate.

Kawamura et al. [2003] found that terrestrial higher plant-derived lipids in Chichijima aerosols are associated with the winter maxima. Similar seasonal variation was reported by *Mochida et al.* [2003b, 2010] for dicarboxylic acids and levoglucosan in Chichijima aerosols. *Chen et al.* [2013] reported saccharides in the aerosols over the island for 1990–1993 and 2006–2009. Although these studies have demonstrated a long-range atmospheric transport of organic aerosols over the remote ocean, there is no study on long-term trends of BB tracers in this region. The purpose of this study is to understand the seasonal and decadal trends of BB tracers over the remote Chichijima Island and identify the mechanisms that control the long-term variations in the WNP. Here we analyzed atmospheric aerosols collected from Chichijima over 13 years from 2001 to 2013 for BB tracers. The data will be discussed in terms of seasonal variations as well as long-term changes of active fire/burning events in Asia and Eurasia. We also discuss the seasonal shifting of atmospheric circulation using BB tracers and based on air mass back trajectories.

2. Materials and Methods

Chichijima is a subtropical island located in the WNP, approximately 2000 km away from the Asian continent and 1000 km south of Tokyo, Japan (Figure 1). The island is too far south to be influenced by the Aleutian Low and too far away from Asia to receive monsoonal rainfall on the equatorward side of the Siberian High. The climate of Chichijima is warm and humid all year round. The ambient temperature varied between 7.8°C and 34.1°C in 1990–1993 [*Kawamura et al.*, 2003].

The detailed aerosol sampling procedure is reported elsewhere [*Chen et al.*, 2013; *Mochida et al.*, 2010]. Briefly, aerosol samples were collected on a weekly basis (January 2001 to November 2013) at the Ogasawara Downrange Station of the Japan Aerospace Exploration Agency on Chichijima Island (27°04′N, 142°13′E, 254 m above sea level). Total suspended particles (TSP) were collected on precombusted (450°C for 6 h) quartz fiber filters (20 × 25 cm, Pallflex 2500QAT-UP) using a high-volume air sampler (Kimoto AS-810A) at a flow rate of 1.0 m^3 min^{-1}. Samples were not collected in November–December 2004 and March–August 2005 due to maintenance at the sampling site.

Punches (21 cm^2) of the filters were extracted with a dichloromethane/methanol (2:1, vol/vol) mixture under ultrasonication. The extracts were filtered through a Pasteur pipet packed with quartz wool to remove filter

debris and then concentrated by the use of a rotary evaporator under vacuum. The extracts were dried with a stream of pure nitrogen gas. The total extracts were reacted with N,O-bis-(trimethylsilyl)trifluoroacetamide with 1% trimethylsilyl (TMS) chloride in the presence of 10 μL of pyridine in a glass vial (1.5 mL) sealed with a Teflon-lined screw cap at 70°C for 3 h to convert the hydroxyl groups of the compounds to the corresponding TMS ethers. The derivatized fractions were diluted with *n*-hexane containing an internal standard of *n*-C_{13} alkane (1.43 ng μL^{-1}) prior to injection into a gas chromatography (GC)-mass spectrometry. More details are presented elsewhere [*Chen et al*., 2013; *Fu et al*., 2008].

Levoglucosan, mannosan, and galactosan were identified by comparing GC retention times and mass spectra with those of authentic standards and literature and library data. Quantification was performed using selected mass fragment ions (e.g., *m/z* 204, 217, and 333) and conversion to compound mass using relative response factors determined by injection of authentic standards. The detailed compound quantification process was adopted from *Fu et al*. [2008]. Recoveries of the target compounds were better than 90%. The data reported here were not corrected for recoveries. Analytical error of the concentrations based on duplicate analysis was generally <15%. Field blanks were collected every 2 months at the sampling site. Target compounds were not detected in the field blanks. The detection limit of levoglucosan was 520 pg μL^{-1}, which corresponds to 0.005 ng m^{-3} for ambient aerosols under a typical sampling volume of 9000 m^3 [*Zhu et al*., 2015]. Levogucosan was used as a surrogate standard for mannosan and galactosan, and the detection limits of the latter compounds were considered to be same as levoglucosan.

Ten-day air mass backward trajectories were calculated every day at 00:00 UTC for each sampling period for 13 years using the NOAA Hybrid Single-Particle Lagrangian Integrated Trajectory (http://ready.arl.noaa.gov/HYSPLIT.php). The starting height of the trajectories is 500 m above the ground. The trajectory-based observations show two major pathways of air masses that arrived in Chichijima. To identify the geographical active fire/burning areas as possible source regions of BB tracers, monthly composite images of the Moderate Resolution Imaging Spectroradiometer (MODIS) active fire/hot spot were obtained from the Earth Observing System Data and Information System (EOSDIS) using the Terra and Aqua satellites (https://earthdata.nasa.gov/data/near-real-time-data/firms/).

3. Results and Discussions

3.1. Concentrations of BB Tracers

We detected levoglucosan (L), mannosan (M), and galactosan (G) as BB tracers in the Chichijima aerosol samples. Their concentrations varied from 0.0003 to 33.6 ng m^{-3}, 0.0002 to 4.80 ng m^{-3}, and 0.0001 to 0.75 ng m^{-3}, respectively. Although their concentrations can significantly fluctuate, L was found as the dominant BB tracer followed by its two isomers: M and G. Figure 2 presents long-term variations of atmospheric concentrations of L, M, and G. Seasonal variations of the BB tracers are characterized by winter/spring maxima and summer/autumn minima, although the concentrations indicate substantial fluctuations. G and M seemingly increased from 2001 to 2006 (Figure 2a). In contrast, L seems to decrease from 2002 to 2009 and then increase toward recent years with the highest concentration in 2013 (Figure 2b), a point to be discussed later. Positive correlations were found for L-M ($r = 0.79$), L-G (0.55), and G-M (0.58). Figure 3 shows box-whisker plots for the concentrations of BB tracers. Median concentrations of BB tracers show a significant increase from 2010 to 2013, although L stayed relatively constant. However, mean concentrations of L seem to increase for the last 4 years.

The previous studies on pelagic sediments from the Pacific Ocean and Chichijima aerosols demonstrated that anthropogenic and biogenic hydrocarbons are transported from the Asian continent to the western and central North Pacific by the westerly winds [*Bendle et al*., 2007; *Kawamura*, 1995; *Ohkouchi et al*., 1997]. *Mochida et al*. [2010] reported that the Chichijima aerosols are characterized by higher abundances of BB organic tracers in the westerly wind regime than in the trade wind regime. Table 1 compares the concentrations of levoglucosan, mannosan, and galactosan in the marine aerosols from Chichijima with those reported in the potential source regions including sites from Sapporo, Japan [*Simoneit et al*., 2004b], Jeju Island, South Korea [*Simoneit et al*., 2004b; *Wang et al*., 2009], Howland Forest Maine [*Medeiros et al*., 2006], Mount Lulin, Taiwan [*Hsu et al*., 2007], Rondônia, Brazil (forest) [*Graham et al*., 2002], Chennai, India [*Fu et al*., 2010], Mount Tai, China [*Fu et al*., 2008], Mount Hua and Mount Tai, China [*Wang et al*., 2012],

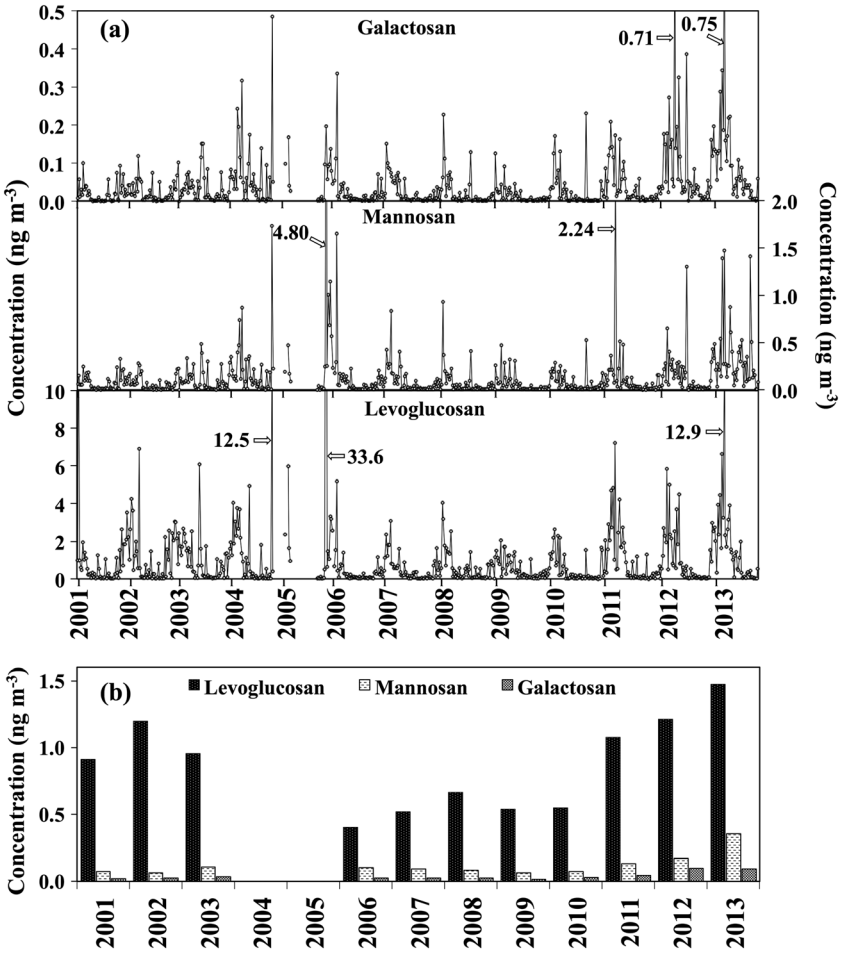


Figure 2. Long-term variations in the concentrations of BB tracers (levoglucosan, mannosan, and galactosan) in the aerosol samples collected at Chichijima for 13 years (2001–2013).

Southeast Asia [*Hu et al*., 2013], and the Northern Hemisphere [*Hu et al*., 2013]. The concentration levels in Chichijima are significantly lower than those reported for continental aerosols from China, India, and Brazil.

The concentrations of BB tracers in the atmosphere are influenced by emission strength of BB from the sources, long-range atmospheric transport, atmospheric circulation, wet and dry deposition, and degradation during the transport. The comparisons of BB tracers between Chichijima and other receptor sites (Table 1), except for a few sites in China and Brazil, indicate that the differences in the concentrations of levoglucosan, mannosan, and galactosan are not very significant when Chichijima is occupied by westerly wind regime. These comparisons suggest that the degradation of levoglucosan during long-range transport is not significant enough to diminish the information of the impact of BB activities to the remote marine atmosphere. The relatively high abundances of levoglucosan reported in the remote regions such as the Arctic Ocean [*Fu et al*., 2009] also suggest the importance of long-range atmospheric transport of BB aerosols and the possible effect of BB on the atmospheric environment in remote receptor sites [*Ding et al*., 2013; *Generoso et al*., 2007; *Warneke et al*., 2010].

3.2. Source Regions of BB-Derived Aerosols Over Chichijima and Active Fire Spots

The climate of Chichijima is influenced by the seasonal exchange of air masses originating from the Pacific Ocean and the Asian continent, which controls the sources and compositions of the aerosols transported to the WNP [*Pavuluri et al*., 2013]. The East Asian monsoon significantly affects the regional climate and air quality in the outflow regions [*Yamamoto et al*., 2011]. The air mass transported to the WNP by the westerly

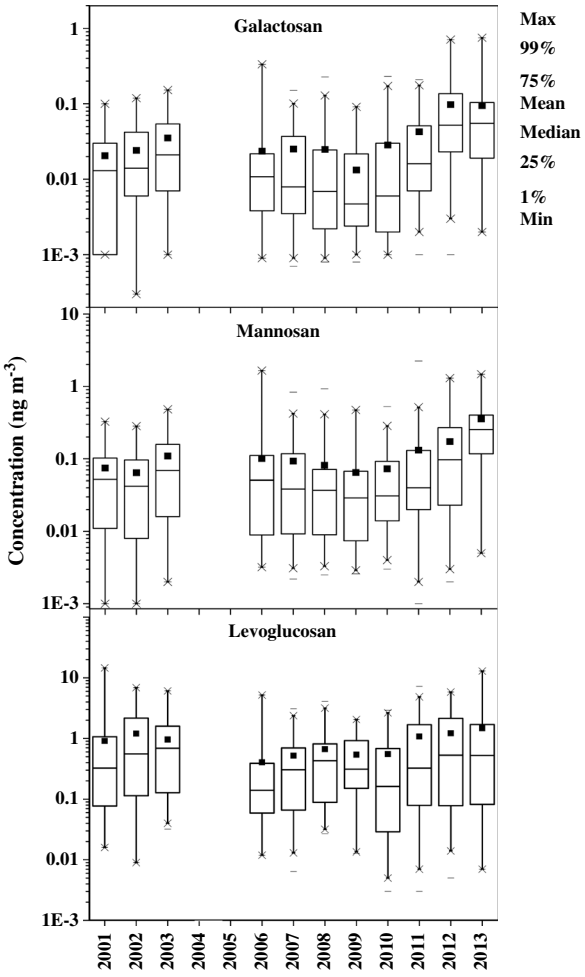


Figure 3. Box-whisker plot of BB tracers (levoglucosan, mannosan, and galactosan) with trends from 2001 to 2013 over Chichijima. The annual average of 2004 and 2005 were excluded due to the significant gap of the data sets.

winds may be enriched with organic aerosols emitted by the forest fires from the Russian Far East and Siberia and the domestic biomass burning in East Asian countries where BB is a common energy source for cooking and space heating.

The above description is consistent with the findings that the Chichijima aerosols are seriously influenced by long-range transport of air masses that are enriched with anthropogenic and natural emissions from Siberia and East Asia in midautumn to midspring [*Kawamura et al*., 2003], although the information of fire/hot spots was not available in the previous study as discussed later. We plotted 13 years of monthly air mass trajectories from 2001 to 2013 to understand the possible changes in the atmospheric circulations (Figures S1a and S1b in the supporting information), but we could not detect any significant year-to-year changes in the atmospheric circulation based on the air mass trajectory analyses. The air mass trajectory analyses indicate that a significant alternate shift between westerly and trade

Table 1. Comparisons of the Concentrations (ng m^{-3}) of Levoglucosan, Mannosan, and Galactosan in Chichijima Aerosols With Those From Different Sites in Asia and Other Regions

Location	Levoglucosan	Mannosan	Galactosan	Reference
Chichijima, western North Pacific (TSP)	0.0003–33.6	0.0002–4.80	0.0001–0.75	This study
Chennai, India (TSP summer)	50.7–213	3.75–20.3	2.71–10.8	*Fu et al*. [2010]
Chennai, India (TSP winter)	4.30–361	0.26–42.6	0.35–23.7	*Fu et al*. [2010]
Mount Tai, China (TSP daytime)	18.5–1730	0.1–61.6	0.02–103	*Fu et al*. [2008]
Mount Tai, China (TSP nighttime)	0.37–3430	0.60–43.1	0.44–46.6	*Fu et al*. [2008]
RondÔnia, Brazil (Forest) ($PM_{2.5}$)	39.9–2660	1.7–127	1.6–44.6	*Graham et al*. [2002]
Howland Forest Maine (TSP)	1.0–55.1	nd [a]–10.4	nd–2.6	*Medeiros et al*. [2006]
Mount Hua, China (nondust storm)	13–106	1.2–30	1.2–9.9	*Wang et al*. [2012]
Mount Hua, China (dust storm period)	25	2.8	3.3	*Wang et al*. [2012]
Mount Tai, China (nondust storm)	0.2–385	0.1–27	0.2–29	*Wang et al*. [2012]
Mount Tai, China (dust storm period)	62–142	7.8–17	7.9–21	*Wang et al*. [2012]
Mount Lulin, Taiwan ($PM_{2.5}$)	1.6–132	nd–0.49	0.06–0.75	*Hsu et al*. [2007]
Gosan, Jeju Island, South Korea (TSP)	8–74	0.2–4.2	nd–3.8	*Simoneit et al*. [2004b]
Sapporo, Japan (TSP)	6.4–56	0.2–15	0.6–2.4	*Simoneit et al*. [2004b]
Southeast Asia (TSP)	1.2–4.3	–	–	*Hu et al*. [2013]
Northern Hemisphere (TSP)	1.1–41	–	–	*Hu et al*. [2013]
Gosan, Jeju Island, South Korea (TSP)	2.8–102	–	–	*Wang et al*. [2009]

[a]nd: below detection limit of ~0.005 ng m^{-3}.

Table 2. Seasonal Variations in the Concentrations (ng m^{-3}) of Biomass Burning Tracers (Levoglucosan, Mannosan, and Galactosan) in Chichijima Aerosols

	Levoglucosan	Mannosan	Galactosan
		Winter	
Range	0.038–14.6	0.006–2.24	0.001–0.75
Mean	2.09	0.23	0.08
Median	1.73	0.14	0.05
SD	1.98	0.31	0.11
		Spring	
Range	0.005–6.89	0.002–1.30	0.0003–0.39
Mean	0.77	0.13	0.04
Median	0.41	0.09	0.02
SD	1.07	0.17	0.06
		Summer	
Range	0.005–1.82	0.0002–1.41	0.0001–0.23
Mean	0.24	0.06	0.02
Median	0.09	0.02	0.01
SD	0.37	0.15	0.03
		Autumn	
Range	0.003–33.6	0.0004–4.80	0.0003–0.49
Mean	0.72	0.11	0.02
Median	0.18	0.02	0.01
SD	2.93	0.43	0.05

wind regimes and thus seasonal atmospheric circulation pattern occurred every year from 2001 to 2013 at Chichijima.

Biomass burning emissions from forest fires, agricultural waste burning, and domestic burning in Asia, the Russian Far East, and Siberia [*Soja et al*., 2007; *Streets et al*., 2003a] may be the possible source of BB tracers detected in the Chichijima aerosols. On the basis of surface fire records and statistics, *Levine et al*. [1995] reported that about 1.5 million hectares of boreal forests are burned annually. The boreal forest fires in Siberia have been suggested to increase in recent years based on ice core analyses of levoglucosan and other BB organic tracers in the Kamchatka Peninsula, Northeast Asia [*Kawamura et al*., 2012]. In addition, tropical deforestation and fires of grasslands, savannas, woodlands, and forests are another important sources of BB tracers.

Figure S2 shows 13 year monthly active fire/hot spot detections in Siberia, East Asia, and South and Southeast Asia. This figure clearly indicates that intensity of active fire/hot spots is lower in winter and higher in late spring to summer [*Lin et al*., 2014]. Even when the forest fire activities were intensive in the Asian regions, they do not contribute to BB tracers over Chichijima in summer (see Figure 2) because there is no air mass transport from the fire-active regions to Chichijima due to the dominant trade winds in summer, except for some cases in Southeast Asia (Figures S1a and S1b). Figures S2a–S2d also demonstrate clear annual variations in the active fire/hot spots with higher intensity in March to April in Southeast Asia. The fire/hot spots seem to increase in recent years (Figures S2a–S2d); another point to be discussed later.

3.3. Seasonal Variations of BB Tracers

Clear seasonal variations were found in the concentrations of BB tracers detected in the aerosol samples from Chichijima. The seasonally averaged concentrations of levoglucosan are significantly higher in winter (2.09 ± 1.98 ng m^{-3}, mean ± standard deviation) than spring (0.77 ± 1.07 ng m^{-3}), autumn (0.72 ± 2.93 ng m^{-3}), and summer (0.24 ± 0.37 ng m^{-3}). The seasonal variations of mannosan and galactosan were similar to those of levoglucosan, although their concentrations are 1 to 2 orders of magnitude lower than levoglucosan (Table 2). The concentrations of BB tracers for some winter/spring samples were very low and comparable to those observed in summer/autumn samples. As depicted in Figure 4a, the large error bars demonstrate that monthly concentrations of BB tracers, except for June to September, significantly fluctuate during the study periods. However, concentrations of BB tracers show a clear seasonal decrease from winter to summer with the order of winter, spring, autumn, and summer (Figure 5).

The concentrations of BB tracers show a significant variation following the seasonal shifts of atmospheric circulation (Figure 4b). The average concentrations of BB tracers are consistent with seasonal changes in air mass trajectories for many years, where BB tracers maximize in January and minimize in September (Figure 4b). The seasonal trend of BB tracers followed the seasonal wind pattern over Chichijima in the WNP, where both wind circulation patterns and concentrations of BB tracers show seasonal variability. Air mass trajectories indicate a clear seasonal shifting with dominant westerlies in winter/spring and trade winds in summer/autumn (Figure 4a). During January to March, the air mass was mainly delivered from the continent, i.e., East Asia, and this flow begins to switch to the Pacific after April. In May to September, the winds arriving at Chichijima come from the central Pacific. Their source regions again shift to the Asian

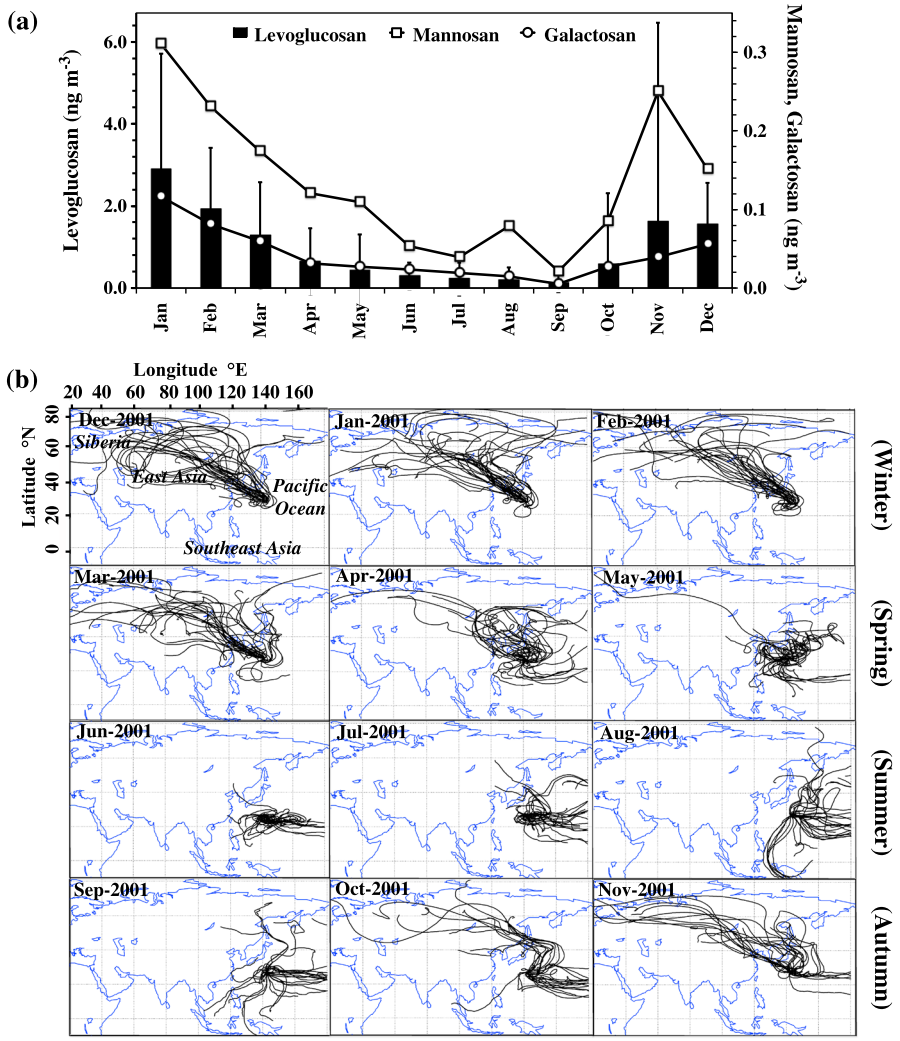


Figure 4. (a) Monthly averaged variations in the concentrations of BB tracers (levoglucosan, mannosan, and galactosan) for 13 years (2001–2013) over Chichijima in the WNP. Error bars indicate the variations (standard deviation) in the concentrations of levoglucosan. (b) Air mass backward trajectories for winter (continental), spring (transition phase, continental to oceanic), summer (oceanic), and autumn (transition phase, oceanic to continental) in 2001.

arid areas from October to December. Concentrations of BB tracers in the Chichijima aerosols are largely controlled by seasonal change in the atmospheric circulation over the WNP.

Maximum concentrations of BB tracers were observed every year in winter/spring. Those peaks should be due to a significant air mass transport from the Asian arid region to the WNP under the influence of strong westerly winds [*Kawamura et al*., 2003], which can carry BB plumes to remote Chichijima Island (Figure 5). *Wang et al*. [2006] also reported levoglucosan in aerosols from 14 Chinese cities, whose concentrations were 3–30 times higher in winter than in summer. The main source of levoglucosan in winter is domestic biomass burning for space heating and cooking in China and other Asian countries [*Streets et al*., 2003b]. The air masses transported by the trade winds are less influenced by the continental outflow and thus are less enriched with BB tracers over Chichijima [*Mochida et al*., 2003b].

Wet deposition is another parameter that controls the atmospheric levels of BB tracers [*Hu et al*., 2013]. However, wet deposition alone could not result in such a large difference of BB tracers in different seasons. In contrast, based on laboratory experiments, *Hennigan et al*. [2010] observed the degradation of levoglucosan by hydroxyl (OH) radicals. It has an average lifetime of 0.7–2.2 days when biomass burning particles are exposed to 1×10^6 molecules cm^{-3} of OH in summer. *Hoffmann et al*. [2010] reported the

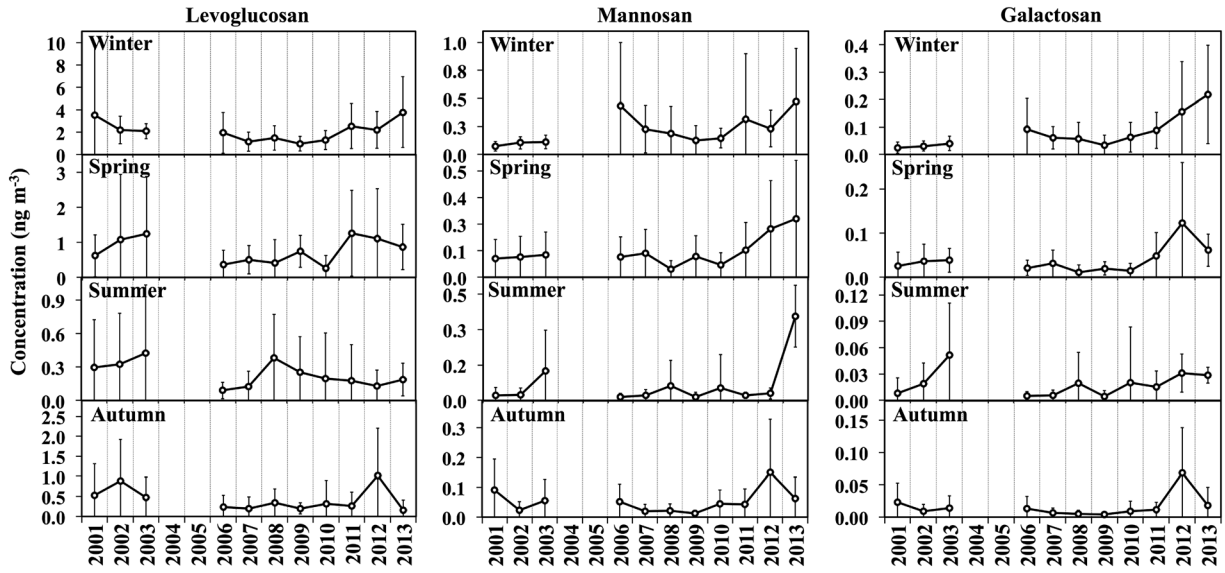


Figure 5. Decadal variations in the seasonal mean concentrations of BB tracers (levoglucosan, mannosan, and galactosan) with seasonal trends from 2001 to 2013. The seasonal means of 2004 and 2005 were excluded due to the significant gap in the data sets. Error bars indicate the standard deviation in the concentrations of BB tracers.

decomposition rate of levoglucosan by OH radicals to be 7.2 ng m^{-3} h^{-1} in summer and 4.7 ng m^{-3} h^{-1} in winter. The levoglucosan-OH reaction intensifies under higher ambient temperature and relative humidity conditions in summer. This may be another cause in lowering the concentrations of levoglucosan in summer, although air masses are only occasionally transported to Chichijima from Southeast Asia where the BB activity is enhanced in summer [*Lelieveld et al.*, 2001; *Pavuluri et al.*, 2010; *Streets et al.*, 2003a]. The summer concentrations of BB tracers may be subjected to photo degradation during atmospheric transport. We consider that the shifts of the air mass origins are the dominant reason for the seasonal variations of BB tracers, while the degradation of BB tracers may notably contribute to low levels of BB tracer in summer.

3.4. Annual Variation of BB Tracers

We found that annual mean concentrations of BB tracers increased toward recent years, especially from 2006 to 2013 (Figures 2 and 3). These increases may be associated with an annual increase of BB activity in the source regions in the Asian continent, followed by atmospheric transport to the WNP. The atmospheric transport from the Asian continent to the North Pacific may also be intensified for the last several years. Interestingly, such an increase in levoglucosan and other BB tracers is consistent with the increased frequency of the active fire/hot spots in the Asian region during the period of 2006 to 2013 (Figures S2a–S2d). The enhanced BB activities in the Asian continent seemingly have more influence on the air quality over Chichijima and the WNP. Meanwhile, concentrations of levoglucosan seemingly declined from 2001–2003 to 2006 although some data are missing for 2004 and 2005. This decline may be associated with decreased fire/hot spots observed by satellite over Asia (Figures S2a and S2b).

Extremely high concentrations of BB tracers were obtained in the sample QFF-2862, which was collected from 31 October to 4 November 2005, with concentrations of levoglucosan to be 33.6 ng m^{-3} and mannosan to be 4.80 ng m^{-3} (Figure 2). Such high concentrations were probably associated with very strong fire events along in the border of northern China, Siberia, and Russian Far East on 25 October 2005 as was documented by MODIS fire spots and smoke plumes (Figure 6a). The 7 day air mass backward trajectories (Figures 6b–6f) also support that the sampling period of QFF-2862 was significantly influenced by air masses associated with source regions where these biomass burning/fire events and smoke plumes were observed.

Although the concentrations of BB tracers showed an increasing trend from 2006 to 2013 as stated above, slightly lower concentrations were detected in 2009 and 2010 (Figures 2 and 3). This might be caused by either a decreased emission rate of BB tracers due to lower BB activities in the source regions or

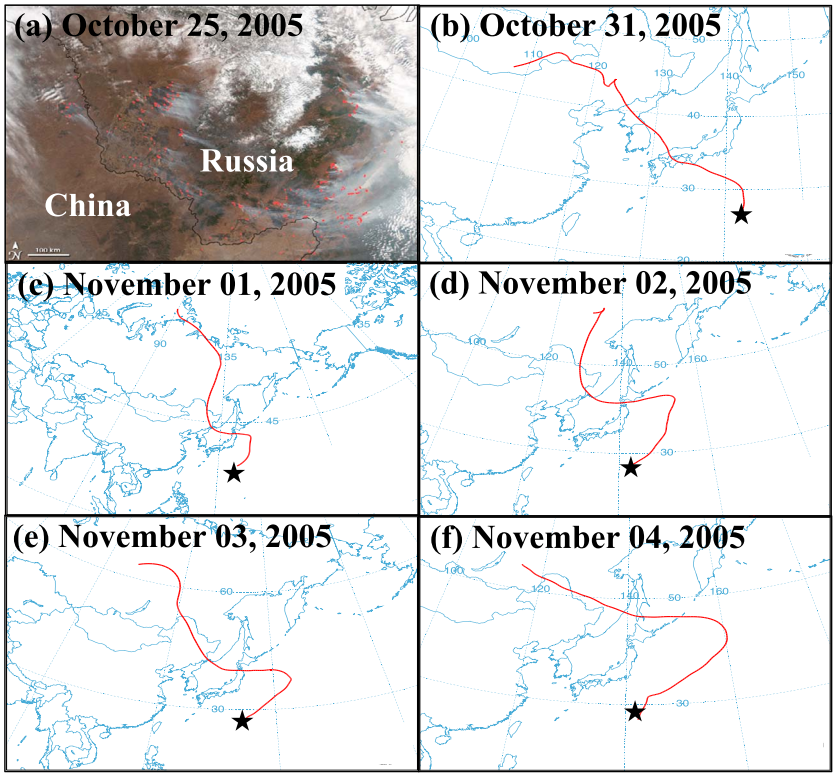


Figure 6. (a) The MODIS fire scores of eastern Russia on 25 October 2005, when biomass burning occurred near the border with northern China, Siberia, and Russian Far East (active fires/burning detected by the sensor are marked in red), (b–f) Air mass back trajectories arriving at Chichijima on 31 October and 1–4 November 2005, respectively (figure site: http://earthobservatory.nasa.gov/Images/).

weakened atmospheric transport during the periods [*Gajović and Todorović*, 2013]. Another possibility is an interannual difference of precipitation. Precipitation events might occur more frequently due to the passage of a low-pressure system during the long-range transport of air masses in winter from the Asian continent [*Wu et al*., 2013; *Zhang and Wu*, 2011]. In fact, we found slightly higher annual precipitation for 2009 and 2010 compared to 2011 at least in Chichijima (Japan Meteorological Agency, http://www.jma.go.jp/jma/indexe.html). The enhanced precipitation scavenges more aerosol particles from the atmosphere, resulting in the lower concentrations of BB tracers in winter over the WNP [*Hu et al*., 2013]. The increase in the wintertime surface air temperature may result in a decrease of domestic BB activities in East Asia, possibly depressing atmospheric transport of BB products over the WNP [*Kim et al*., 2013; *Yang and Wu*, 2013].

By comparing the annual concentrations of BB tracers in Chichijima aerosols with the MODIS active fire/hot spots, we found a strong similarity between the two parameters. As seen in Figures 2a and 2b, the concentrations of BB tracers substantially increase from 2009 to 2013. It is important to note that the BB emissions due to forest fires in Siberia and the Russian Far East as well as residential wood combustion and agricultural waste burning in the East Asian countries increased over the last decade [*Gajović and Todorović*, 2013; *Generoso et al*., 2007; *Ito*, 2011]. This phenomenon implies that more BB products in the source regions are being emitted to the air and transported over the WNP in the winter/spring periods under westerly wind conditions [*Kawamura et al*., 2003; *Mochida et al*., 2003b, 2010].

It is noteworthy that the length of the high-concentration period also significantly increased from 2006 to 2013 (Figure 2). Further, the active fire/hot spot images show an enhancement for these years, being consistent with the increase in the observed concentrations of levoglucosan and other BB tracers (Figures 2 and 3). We found positive linear relations between annual mean concentrations of levoglucosan ($r = 0.54$, $p = 0.17$), mannosan ($r = 0.52$, $p = 0.19$), and galactosan ($r = 0.61$, $p = 0.11$) in Chichijima aerosols

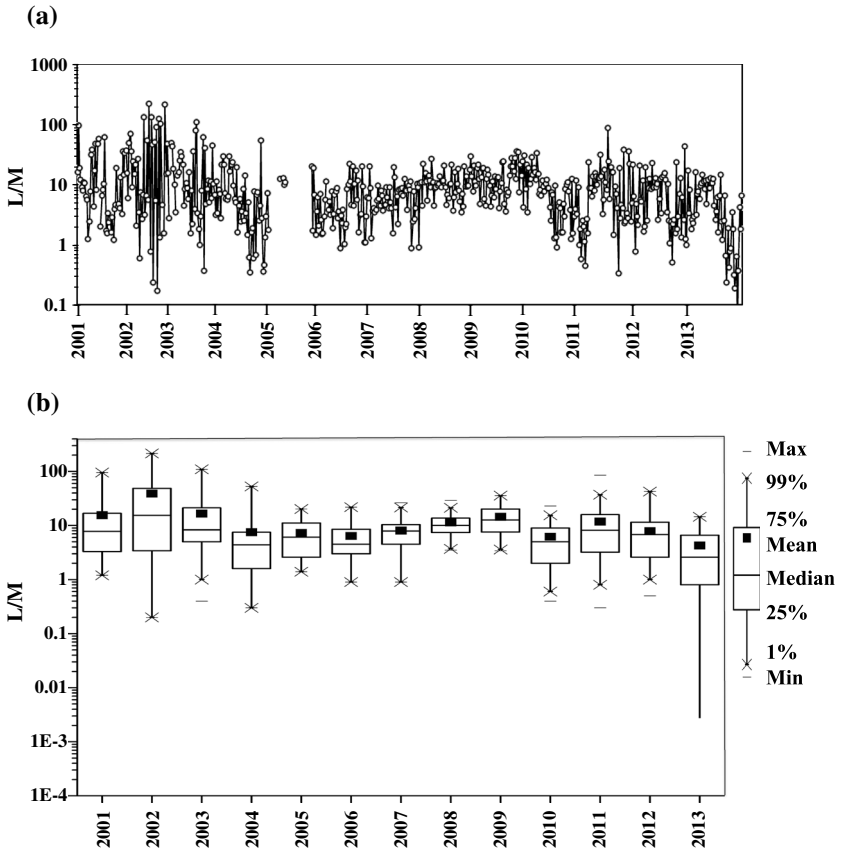


Figure 7. (a) Log plot and (b) box-whisker plot of levoglucosan to mannosan ratios (L/M) in the Chichijima aerosol samples for 2001 to 2013.

and fire counts for the region of 30–70°N and 80–140°E over 2006–2013 (Figures S3a–S3d). The periods of high fire/hot spot frequency increased from 2006 to 2013 (Figures S2a–S2d), indicating that the BB activities are recently increasing in the East Asian regions [*Gajović and Todorović*, 2013], which may be associated with the fire activities under drier conditions. This may be associated with the recent increase of wintertime ambient temperature in Southeast Asia [*Yang and Wu*, 2013] and annual temperature in Siberia [*Konya et al*., 2014], which may cause drier condition and thus enhance fire activities. Air mass trajectories further support that the air masses had passed through Siberia, East Asia, and the Russian Far East before arriving at Chichijima (Figure 4b and Figures S1a and S1b). Thus, BB activities in the source regions of the Asian continent primarily have a significant impact on aerosol compositions in the marine atmosphere over the WNP during the winter/spring period.

3.5. Levoglucosan/Mannosan Ratio

We calculated levoglucosan/mannosan (L/M) ratios to distinguish the possible source difference of BB tracers in the Chichijima aerosol samples. Figures 7a and 7b represent the 13 year variation of L/M ratios and yearly averaged variations, respectively. Interestingly, L/M ratios are found to be higher in summer/autumn and lower in winter/spring. We also found very high L/M ratio in 2002 (mean 36) followed by 2001 (16), 2003 (16), 2009 (15), and 2004 (12). The seasonal L/M ratios (summer 55, autumn 52, and winter 32) were also observed to be higher in 2002. They are much higher than those reported for urban aerosols (summer 6.4 and winter 7.8) from Belgium [*Pashynska et al*., 2002] and those (summer 5.5 and winter 4.7) from Ireland [*Kourtchev et al*., 2011]. Higher springtime L/M ratios (15–40) were reported for Asian aerosol at Gosan, Jeju Island, South Korea [*Simoneit et al*., 2004b]. High L/M ratio (40) has been reported by *Engling et al*. [2013] for south central Taiwan during the summer of 2007. In our study, the long-term variation of L/M ratios in

the Chichijima aerosols show the range of 0.03 to 216 (12.2 ± 19.6, median 7.5). These L/M ratios are within those (21.7 and 100) reported by *Iinuma et al*. [2007] and *Engling et al*. [2006], respectively, for African savanna grass. The L/M ratios of 10–13 for particulate matter ($PM_{2.5}$) and of 9–13 for PM_{10} were reported in Tanzania by *Mkoma and Kawamura* [2013], which are also close to our values.

Several studies suggested that L/M ratios could be used for discriminating hard wood (angiosperm) and soft wood (gymnosperm) burning [*Kawamura et al*., 2012; *Kuo et al*., 2011]. Hard wood contains higher amount of cellulose (55–65%) than hemicellulose (20–30%) [http://bioenergy.ornl.gov/main.aspx; *Klemm et al*., 2005; *Sjostrom*, 1993]. Levoglucosan is a thermal decomposition product of cellulose, whereas mannosan and galactosan are derived by thermal decomposition of hemicellulose [*Simoneit*, 2002]. Mannosan and galactosan are thermally less stable than levoglucosan [*Simoneit*, 2002]. Higher L/M ratios reflect hard wood burning and lower ratios for soft wood burning. In a laboratory chamber study, *Schmidl et al*. [2008] observed L/M ratios of 3–5 for soft wood and 14–15 for hard wood burning. *Engling et al*. [2009] reported >10 and 3–5 for L/M ratios for hard wood and softwood, respectively. They also reported L/M ratios up to 50 for some herbaceous plants.

The ratios reported by *Sheesley et al*. [2003] and *Zhang et al*. [2007] for rice straw (41.6) and cereal straw (55.7) burning, respectively, are also lower than the high ratios (>100) observed in our study. In addition, a recent study by *Kuo et al*. [2011] suggested that the burning temperature plays a significant role in the emission of levoglucosan and mannosan; that is, their concentrations increased with an increase in the burning/combustion temperature up to 250°C and 200°C, respectively. The mannosan production declines at a temperature >300°C. The effect of burning and combustion temperature on the emission of levoglucosan and mannosan were also discussed in *Engling et al*. [2006]. High emission occurred during high-temperature flaming as opposed to low-temperature smoldering combustion [*Gao et al*., 2003; *Iinuma et al*., 2007].

Based on the above discussions, we can hypothesize that higher summertime L/M ratios in 2002 followed by 2001 and 2003 were due to forest fires characterized by high-temperature flaming combustion in Southeast Asia, which should emit levoglucosan-enriched biomass burning aerosols. At higher temperatures, mannosan preferentially decomposes [*Engling et al*., 2013]. The higher L/M ratios in 2002 (Figure 7b) may be associated with enhanced biomass burning in Southeast Asia, as supported by fire/hot spots (Figure S2). In section 3.2, we already discussed the occasional air mass transport from Southeast Asia to Chichijima in summer, which delivers levoglucosan-enriched biomass burning aerosols. This might be the possible reason why higher L/M ratios were observed in summer than in winter.

Furthermore, there are several studies that have reported enhanced summertime forest fires in North America and Canada [*Dibb and Jaffrezo*, 1997; *Kehrwald et al*., 2012]. Chichijima Island is highly influenced by trade winds during summer and autumn. It could be assumed that the air masses are transported from North America and Canada, which might be another source region for high L/M ratios in Chichijima aerosols. Although back trajectories seemingly do not to support the above idea because even 10 day back trajectories cannot reach the continent (North America) in summer/autumn, this pathway could not be eliminated. The photochemical decomposition of levoglucosan during summer [*Hennigan et al*., 2010; *Hoffmann et al*., 2010] may contribute to trace levels of levoglucosan in Chichijma ($0.2\ ng\ m^{-3}$) after long-range transport of levoglucosan-enriched air masses possibly from North America [*Fraser and Lakshmanan*, 2000; *Genualdi et al*., 2009; *Simoneit et al*., 1999].

In spite of the above discussions, the high L/M ratios in summer/autumn aerosols from Chichijima cannot be clearly explained, although there are few studies that reported very high L/M ratios (>40). For example, burning products of rice and cereal straws by chamber experiments show higher L/M ratios of 40–55 [e.g., *Engling et al*., 2009]. Although the field burning of these straws likely occurs in summer/autumn in East Asia, the straws are also used in winter for house heating and cooking in China. *Fu et al*. [2008] reported high L/M ratio (46) for biomass burning plumes derived from field burning of wheat straw in the North China Plain. Further, very high L/M ratios (30–90) are reported for lignite burning at low temperature (200°C) [*Kuo et al*., 2011]. Because lignite is used for space heating and cooking in China, such burning products may contribute via long-range atmospheric transport to the high L/M ratios that are often observed in the Chichijima aerosols (Figure 7). It is of importance to further search the sources of high L/M ratios in future studies.

4. Summary and Conclusions

Based on 13 years of observations on marine aerosols from Chichijima in the WNP, we found increased concentrations of biomass burning (BB) organic tracers (e.g., levoglucosan) in winter/spring, which are mainly transported from East Asia by the westerly winds. The seasonal and annual loading of BB tracers were significantly controlled by atmospheric circulation, which is associated with the delivery of continental and anthropogenic air masses enriched with BB tracers to the WNP by the westerly winds in midautumn to midspring. Although the air masses during midspring to midautumn are less influenced with BB, air masses that contain BB tracers are transported occasionally from Southeast Asia over the WNP in July/August. Seasonal variations in the concentrations of BB tracers in Chichijima aerosols are caused by a seasonal shift of atmospheric circulation over the WNP. We found that the concentrations of BB tracers increased from 2006 to 2013 and that the high-concentration periods were also prolonged in those years due to the continuous increase of BB activities in East Asia as supported by fire spot images. The long-term L/M ratios that showed higher values in summer may suggest the occurrence of high-temperature burning in Southeast Asia and/or differences in biomass compositions, followed by vigorous transport of levoglucosan-enriched BB products over Chichijima Island. This study suggests that the long-range transport of BB products to the WNP may potentially cause an impact on climate from a regional to global scale because BB products are hygroscopic and thus have an influence on the radiative forcing of aerosols by acting as cloud condensation nuclei.

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