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LETTER

Exceedances of air quality standard level of $PM_{2.5}$ in Japan caused by Siberian wildfires

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Abstract

We revisited long-term observations of $PM_{2.5}$ at ground-based stations in Japan during 2001–2012 to examine possible impacts of Siberian wildfires on regional air quality. Exceedances of Japan's air quality standard for daily mean concentration (35 $\mu g\ m^{-3}$) were observed several times at Rishiri Island in northern Japan in the spring of 2003 and 2008 when intense wildfires occurred in Siberia. Satellite observations showed that aerosols and CO originating from biomass burning were transported from Siberia toward Japan. The regional chemical transport model also demonstrated that the $PM_{2.5}$ enhancements during high $PM_{2.5}$ days (>35 $\mu g\ m^{-3}$) were attributed to Siberian wildfires, suggesting that the contribution from Siberian biomass burning had a critical impact on exceedances of air quality standard level. The monthly (May) and annual mean $PM_{2.5}$ concentrations in 2003 were about twice and 20% higher, respectively, than those of the long-term average at Rishiri Island, where the influence of Siberian wildfires was the largest in Japan. Except for 2003 and 2008, a high $PM_{2.5}$ day due to Siberian wildfires was not identified. Although Siberian biomass burning does not affect the air quality standard of $PM_{2.5}$ for the years without strong fires, it causes exceedance of the air quality standard level when intense fires occur.

1. Introduction

Emissions from vegetation fires are a significant source of trace gases and atmospheric aerosols. Aerosol particles and precursor gases of ozone (O_3) such as NO_X, volatile organic compounds (VOCs), and carbon monoxide (CO) emitted from vegetation fires affect air quality and climate (Langmann *et al* 2009). Aerosols (also known as particulate matter) are recognized as one of the most important air pollutants. $PM_{2.5}$ (particulate matter with a diameter of 2.5 μm or less) has drawn much attention because exposure to high-concentration $PM_{2.5}$ can have adverse effects on human health (US Environmental Protection Agency 2009). Since observational studies based on satellite data, airborne measurements, and ground-based measurements and model simulations indicate that aerosols caused by fires can be transported on regional and intercontinental scales (e.g. Damoah *et al* 2004, Bertschi and Jaffe 2005, Lee *et al* 2005, Stohl *et al* 2006), it is a significant issue to estimate the influence of particulate pollution due to boreal fires on air quality in the downwind regions.

Siberian wildfires are strong emission sources, which significantly contribute to global biomass burning emissions (van der Werf *et al* 2010). Siberian forests experience wildfires from spring to autumn every year, but their activities show a considerable interannual variability (van der Werf *et al* 2010, Tanimoto *et al* 2015). For example, intense fires occurred across the Siberian forests in 2003, which caused air quality degradation in distant regions. Elevated concentrations of O_3, CO and aerosols originating from Siberian wildfires were observed over Northeast Asia (Lee *et al* 2005, Kaneyasu *et al* 2007, Jeong *et al* 2008, Tanimoto *et al* 2008, Tanimoto *et al* 2009). It was also reported that air pollutants were transported from Siberia to North America across the Pacific (Jaffe *et al* 2004, Bertschi and Jaffe 2005) and to the Arctic region (Generoso *et al* 2007).

In this study, we investigated the influence of biomass burning in Siberia on $PM_{2.5}$ pollution in Japan

during 2001–2012 through ground-based measurements, satellite observations, and a regional chemical transport model. In Japan, an air quality standard for $PM_{2.5}$ was newly introduced in 2009, with an annual mean value of 15 $\mu g\,m^{-3}$ and a daily mean value of 35 $\mu g\,m^{-3}$. Recently, Ikeda *et al* (2015) estimated the contributions from various source regions in East Asia to $PM_{2.5}$ over Japan, but they focused on anthropogenic emissions, and thus contributions from natural sources including wildfires are not yet investigated. Whereas East Asia is the most significant source region of anthropogenic air pollutants in global emissions, it also has important natural emission sources of $PM_{2.5}$ such as Siberian biomass burning and dust, and these can affect the atmospheric environment in this region. Previous studies dealing with intense wildfires in Siberia investigated enhancements of aerosols (aerosol optical depth and PM_{10}) as well as O_3 and CO, but their influences on air quality standard for $PM_{2.5}$ concentration has not been examined. Since the fire activities in boreal forests are predicted to increase under a future warming climate (Stocks *et al* 1998, Malevsky-Malevich *et al* 2008, Tchebakova *et al* 2009), it is important to investigate the influence of Siberian wildfires based on long-term data to obtain a better perspective for their impacts on future air quality. In this letter, we show that high $PM_{2.5}$ concentrations exceeding the Japanese air quality standard for the daily mean value (35 $\mu g\,m^{-3}$) were observed at Rishiri Island in northern Japan during intense fire seasons in 2003 and 2008. Both satellite observations of aerosols and CO and model simulations demonstrated that the exceedances of the air quality standard were caused by long-range transport from Siberian wildfires.

2. Observation data and model simulation

2.1. Observation data

Observational data of $PM_{2.5}$ mass concentrations used in this study were obtained at Rishiri Island (45.11°N, 141.20°E) and Oki Islands (36.28°N, 133.18°E) by the Acid Deposition Monitoring Network in East Asia (EANET), and Nonodake (38.55°N, 141.17°E) by the Japanese Ministry of the Environment. Hourly CO data were also available at Rishiri (Tanimoto *et al* 2009). We used aerosol optical depth (AOD) data at a wavelength of 550 nm from the Moderate Resolution Imaging Spectroradiometer (MODIS) aboard the Terra and Aqua satellites. The MODIS Terra and Aqua AOD data were measured at the local equatorial overpass time of about 10:30 am and 1:30 pm, respectively. The MODIS data used in this study were the daily and monthly level-3 AOD products of Collection 5.1 with a horizontal resolution of 1° × 1°. We used CO total column data observed by the Atmospheric Infrared Sounder (AIRS) on board the Aqua satellite. The AIRS CO data used were the level-3 daily products (version 6) of the ascending orbit gridded on a horizontal resolution of 1° × 1°.

2.2. Model description

We used the Weather Research and Forecasting (WRF) model version 3.3.1 (Skamarock *et al* 2008) to simulate meteorological fields for a chemical transport model. The model domain was centered at 37°N, 115° E on the Lambert conformal projection, covering East Asia and Siberia (figure 2). The model used a horizontal grid resolution of 80 km with 98 × 94 grid points. The vertical layers consisted of 38 levels from the surface to 50 hPa. The initial and boundary conditions were obtained from the National Center for Environmental Prediction (NCEP) Final Operational Global Analysis (FNL, ds083.2) data (six-hourly, 1° × 1° resolution). In the model domain, three-dimensional grid nudging was used for horizontal wind, temperature, and the water vapor mixing ratio every six hours. We used the following parameterizations: the Kain-Frisch scheme for cumulus parameterization, the WRF Single-Moment 6-Class Microphysics (WSM6) scheme for microphysical parameterization, the Goddard scheme and the Rapid Radiative Transfer Model (RRTM) for shortwave and longwave radiation processes, and the Mellow-Yamada-Nakanishi-Niino level-2.5 scheme for planetary boundary layer parameterization.

The Community Multi-scale Air Quality model (CMAQ) version 4.7.1 (Byun and Schere 2006) was used as a chemical transport model in this study. The SAPRC99 (Statewide Air Pollution Research Center, Version99, Carter 2000) scheme was used for the gas-phase chemistry. Aerosol processes were simulated by the fifth generation CMAQ aerosol module (AERO5). The AERO5 uses ISORROPIA (Nenes *et al* 1998) as the thermodynamic equilibrium module of inorganic aerosols. For a secondary organic aerosol model, the scheme developed by Carlton *et al* (2010) is incorporated. The aerosol size distribution is represented by three lognormal distributions: Aitken, accumulation, and coarse modes (Binkowski and Roselle 2003). We calculated the $PM_{2.5}$ mass concentration as the total mass of the Aitken and accumulation modes of SO_4^{2-}, NO_3^-, NH_4^+, Na^+, Cl^-, elemental carbon (EC), primary and secondary organic aerosols, and primary $PM_{2.5}$. The initial and boundary conditions were derived from the CMAQ default data.

Anthropogenic emission data were derived from the Regional Emission inventory in Asia (REAS) version 2.1 (Kurokawa *et al* 2013). We used the Global Fire Emission Database (GFED) version 3.1 (van der Werf *et al* 2010) for daily emissions from biomass burning. The GFED inventory provides gaseous species (NO_X, CO, VOCs, SO_2, and NH_3) and primary aerosol emissions (organic carbon (OC), black carbon (BC), and $PM_{2.5}$). For primary $PM_{2.5}$ emissions, difference between $PM_{2.5}$ and the sum of BC and OC was

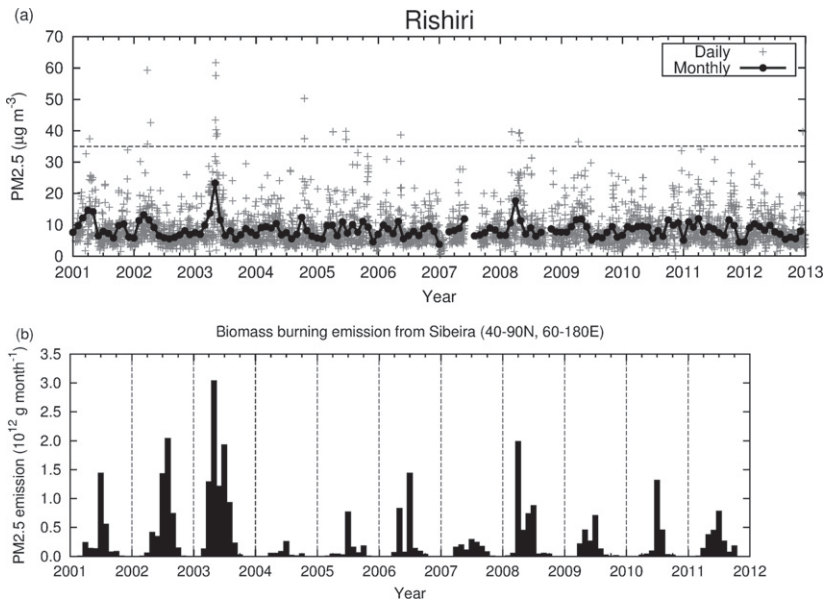

Figure 1. (a) Temporal variations of daily and monthly mean $PM_{2.5}$ mass concentrations at Rishiri for 2001 to 2012. Dashed line indicates Japan's air quality standard for daily mean concentration (35 $\mu g\ m^{-3}$). (b) Monthly biomass burning emissions of primary $PM_{2.5}$ in Siberia (40°N–90°N, 60°E–180°E) obtained from GFEDv3.1.

used as unspecified $PM_{2.5}$. Biomass burning emissions were distributed from the surface to 1000 m in the model. Biogenic emission data were taken from the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2 (Guenther *et al* 2006). Volcanic emission data for SO_2 were based on Streets *et al* (2003). In addition to the standard simulation with biomass burning emissions, we performed a sensitivity simulation in which emissions from biomass burning in Siberia (>40°N) were excluded to quantify the impact of biomass burning in Siberia on $PM_{2.5}$ mass concentrations. We considered the difference of simulated $PM_{2.5}$ concentration between the sensitivity run and the standard simulation with unchanged emissions as the contribution from Siberian wildfires. The model simulations were conducted for severe fire seasons in 2003 and 2008 to evaluate the contribution from Siberian wildfires to $PM_{2.5}$ concentrations. The simulation periods were from April 1 to May 31 2003 and from March 1 to May 31 2008, and the first month of each simulation was used for model spin-up.

3. Results and discussion

Figure 1 shows the temporal variations of daily and monthly mean $PM_{2.5}$ mass concentrations at Rishiri Island from 2001 to 2012. Rishiri Island is a remote island off the coast of Hokkaido in northern Japan, and the influence of local pollution is negligible. The observed $PM_{2.5}$ concentrations in northern Japan are generally low compared to western Japan because of a longer distance from the Asian industrial source regions, and the annual mean concentration at Rishiri was the lowest among the observation sites in Japan for the year 2010 (Ikeda *et al* 2015). Actually, monthly mean concentrations at Rishiri were basically less than 10 $\mu g\ m^{-3}$ during the period of 2001–2012, but the daily mean values showed occasional peaks and which exceeded even the air quality standard (35 $\mu g\ m^{-3}$). Daily mean $PM_{2.5}$ concentrations exceeding the air quality standard were observed 1–6 times a year for 2001–2006, 2008–2009, and 2012 (table 1). Exceedances of air quality standard occurred mainly in spring, except in 2004, 2005, and 2012, when they occurred in autumn, summer and winter, respectively. The numbers of high $PM_{2.5}$ days (>35 $\mu g\ m^{-3}$) were 6 in the spring of 2003 and 4 in 2008, and the monthly mean concentrations were clearly higher in May 2003 (23.3 $\mu g\ m^{-3}$) and April 2008 (17.7 $\mu g\ m^{-3}$) than in other months. These months correspond to the periods during which intense wildfires occurred in Siberia (figure 1(b)), suggesting the influence of long-range transport from Siberian biomass burning. For high $PM_{2.5}$ days in the spring of 2001, 2002, and 2005, dust was detected by visibility-based observations at the meteorological stations in Hokkaido (Wakkanai and/or Asahikawa) near Rishiri Island. Thus the increases in $PM_{2.5}$ concentrations during these days (e.g. 11 April 2001 and 22–23 March 2002) were probably influenced by the dust. Next, we examine the impact of $PM_{2.5}$ originating from biomass burning in Siberia on the exceedances of the air quality standard for daily mean $PM_{2.5}$ concentration in the spring of 2003 and 2008 using satellite measurements and model simulations.

Figure 2 shows the horizontal distribution of monthly biomass burning emission of primary $PM_{2.5}$ obtained from GFEDv3.1 in May 2003 and April 2008.

Table 1. Number of high $PM_{2.5}$ days exceeding the air quality standard for daily mean concentrations in Japan (35 $\mu g\,m^{-3}$)[a], annual mean concentrations, and primary $PM_{2.5}$ emissions (Tg $year^{-1}$) from Siberian biomass burning (40°N–90°N, 60°E–180°E) obtained from GFEDv3.1[b].

Year	Number of high $PM_{2.5}$ days	Annual mean concentration ($\mu g\,m^{-3}$)	Primary $PM_{2.5}$ emission (Tg $year^{-1}$)
2001	1 (1)	9.0	2.7
2002	3 (3)	8.0	5.3
2003	6	9.8	8.8
2004	2	8.2	0.5
2005	3 (1)	8.2	1.3
2006	1	8.1	2.7
2007	0	7.6	1.3
2008	4	9.1	4.3
2009	1	8.4	1.8
2010	0	8.6	2.0
2011	0	8.5	2.3
2012	1	7.5	–

[a] Values in brackets denote the number of days when dust was detected by visibility-based observations at the meteorological stations near Rishiri Island (Wakkanai and/or Asahikawa) for high $PM_{2.5}$ days.

[b] GFEDv3.1 data were not available in 2012.

Intense emissions occurred in the east of Lake Baikal and along River Amur, the border between Russia and China. The emissions of primary organic carbon (POC) and BC from Siberia (40°N–90°N, 60°E–180°E) during May 2003 were estimated to be 2.1 and 0.15 Tg, which were approximately five times higher than the long-term average in May from 2001 to 2010. The POC emission from the Siberian biomass burning in May 2003 was comparable to that of the anthropogenic emissions over East Asia for 2003 (3.0 Tg y^{-1}) in REASv2.1 (Kurokawa *et al* 2013). In April 2008, the intense emission regions were similar to those in May 2003, but the emissions along River Amur were stronger compared with the Baikal region. The Siberian biomass burning emissions of POC and BC were estimated to be 1.3 and 0.10 Tg in April 2008, about 30% less than those in May 2003.

Figure 3 shows the temporal variations of the observed and modeled daily mean $PM_{2.5}$ mass concentrations and CO mixing ratio at ground-based measurement sites over Japan in May 2003 and April 2008. The most significant elevation of $PM_{2.5}$ concentrations was observed at Rishiri, located in the most northern part of Japan. The observed results at Rishiri showed enhancements of $PM_{2.5}$ concentrations on 1–3, 4–6, 7–8, 11–13, and 15–16 May 2003. The daily mean $PM_{2.5}$ concentrations on 4–7 and 14 May were 42.4–62.3 and 40.7 $\mu g\,m^{-3}$, respectively, which exceeded the air quality standard value for 24 h in Japan (35 $\mu g\,m^{-3}$). The observed CO mixing ratio at Rishiri obviously showed increases at the same time as $PM_{2.5}$, reaching a maximum of 340 ppbv on 5 May. The enhancements of $PM_{2.5}$ concentrations at Nonodake and Oki were not so significant as those at Rishiri, but relatively high concentration events over 35 $\mu g\,m^{-3}$ were also observed at these sites. At Nonodake, the observed $PM_{2.5}$ concentration exhibited moderate elevations on 1–7 and 8–15 May, peaking at 34 and 33 $\mu g\,m^{-3}$, respectively. The temporal variations of the observed $PM_{2.5}$ concentration at Oki showed an increase with a maximum of 37 $\mu g\,m^{-3}$ on 24–25 May in addition to a moderate event in early May.

The model generally reproduced the temporal variations of $PM_{2.5}$ concentrations including almost all of the elevated events in May 2003. The correlation coefficients of daily mean concentrations between the observations and the model results were 0.81, 0.56, and 0.77 at Rishiri, Nonodake, and Oki, respectively. For the event during 4–6 May at Rishiri, the standard simulation showed a $PM_{2.5}$ enhancement consistent with the observations. By contrast, the concentration of the sensitivity simulation without biomass burning emissions from Siberia did not increase and remained very low over the entire period of this event. This difference between the simulations indicated that the significant $PM_{2.5}$ pollution for these three days was caused mostly by the wildfires in Siberia. Although the maximum concentration of the simulated $PM_{2.5}$ was smaller than that of the observation, this feature was also found for the event on 14–15 May at Rishiri, demonstrating the dominant contribution from Siberian biomass burning. These model results suggest that $PM_{2.5}$ originating from Siberian wildfires had a substantial impact on air quality, exceeding the standard daily mean values on 4–7 and 14 May 2003. During 1–3 May at Rishiri, the concentration of the sensitivity simulation increased from 2 May and the difference of concentrations between the simulations with and without Siberian biomass burning emissions was small in contrast to the early part of the event, suggesting a large contribution other than biomass burning in Siberia. The increase during the late part of the event could be attributed to $PM_{2.5}$ originating from anthropogenic emissions in the Asian continent (Ikeda *et al* 2014, Ikeda *et al* 2015). The model also captured enhancements of CO at Rishiri, but tended to underestimate the observed results. The CO mixing ratio used in the western boundary condition of 80 ppbv was lower than that of the AIRS observations (~150 ppbv), probably causing an underestimation by the model. Uncertainties in the biomass burning emission inventory might also result in these negative biases (e.g. Dolman *et al* 2012, Konovalov *et al* 2014). The concentrations in the sensitivity simulation without biomass burning in Siberia showed little increase during the elevated events except for 2−3 May. Thus, the model simulations suggest that the enhanced CO at Rishiri was also attributed to Siberian biomass burning. In addition to Rishiri, the influences of Siberian wildfires on $PM_{2.5}$ were seen at Nonodake and Oki located in eastern and western Japan, respectively. The

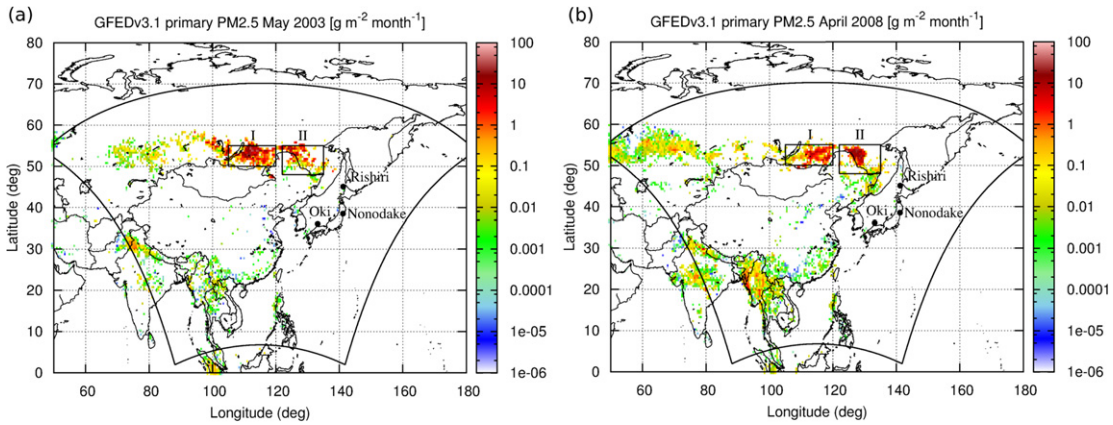

Figure 2. Horizontal distributions of primary $PM_{2.5}$ emissions from biomass burning obtained from GFEDv3.1 in May 2003 (a) and April 2008 (b). Model domain is shown by solid line. Black circles indicate locations of ground-based measurement sites. Rectangles I and II denote major fire areas in Siberia.

model results showed that the $PM_{2.5}$ enhancements on 1–2 and 9–13 May at Nonodake were largely affected by biomass burning in Siberia. At Oki, contributions from Siberian wildfires were found during the events on 9–10 and 23–25 May.

Figure 4 shows the horizontal distributions of simulated surface $PM_{2.5}$ and AOD from MODIS observations averaged for 4–6 May 2003, when a significant $PM_{2.5}$ pollution event with daily mean concentrations over 35 $\mu g\,m^{-3}$ occurred at Rishiri (figure 3(b)). The concentrations of simulated $PM_{2.5}$ were the highest around the regions where biomass burning emissions were large, such as east of Lake Baikal and along the Amur River (figure 2(a)). Polluted plumes spread eastward from the source regions by westerly winds that prevailed over eastern Siberia. The model simulation demonstrated that the high-$PM_{2.5}$ air masses were transported across northeast China and Maritime Province of Siberia, and reached the northern part of Japan. In contrast to high latitudes (>45°N), easterly winds were predominant in central east China and over the Yellow Sea, the East China Sea, and the Sea of Japan, and thus they could prevent the spread of the polluted continental air mass to the west during this period. The AOD from MODIS observations showed high values from northeast China to northern Japan and the western Pacific. Considering the circulation pattern as described above, these areas corresponded to the downwind region of Siberian wildfires, and thus it can be suggested that the enhanced AOD was mainly attributed to aerosols originating from biomass burning in Siberia.

The observed and simulated CO total column averaged for 4–6 May are also shown in figure 4. The AIRS observation showed elevated value of CO from east of Lake Baikal to the northwestern Pacific, particularly northeast China. This spatial pattern was similar to that of AOD from MODIS observations, suggesting that CO emitted from Siberian wildfires was transported to northern Japan along with aerosols, as observed at the surface of Rishiri (figures 3(a), (b)). The model simulation well reproduced the horizontal distribution of observed CO total column, but the values were lower than those of the AIRS observations. Possible reasons for the underestimation by the model are due to the boundary condition of CO used for the CMAQ simulation and uncertainties in biomass burning emissions as mentioned above. The simulated enhancements were more apparent over burning locations compared with the AIRS observations, which exhibited larger CO enhancements in the downwind region of the intense emissions. This discrepancy is probably because AIRS has little sensitivity to the boundary layer CO and detects CO enhancements mainly in the middle troposphere (Tanimoto *et al* 2009).

The exceedance of air quality standard for daily mean $PM_{2.5}$ concentration due to dominant contribution from Siberian biomass burning was also seen in April 2008. On 26 April, the daily mean concentrations at Rishiri and Nonodake were 39.4 and 37.7 $\mu g\,m^{-3}$, respectively (figures 3(f), (g)). The simulation with biomass burning emissions in Siberia could reasonably reproduce these enhancements well. In April 2008, correlation coefficients of daily mean concentrations were 0.85–0.95. The sensitivity simulation without emissions from Siberian biomass burning showed little increase in $PM_{2.5}$ concentration. Thus, the elevations during this event were mostly attributed to $PM_{2.5}$ originating form Siberian wildfires. The daily mean concentrations also exceeded on 21 April and 1 May at Rishiri. However, the concentration of sensitivity simulation increased during these events, suggesting that sources other than Siberian biomass burning had a substantial contribution to these enhancements.

Next, we discuss the model reproducibility in the two intensive fire years of 2003 and 2008. Because Rishiri Island is situated very close to and just downwind of far Eastern Siberia, the comparison at this site is simple relative to those at further downwind sites, and provides important implications for evaluating and improving the

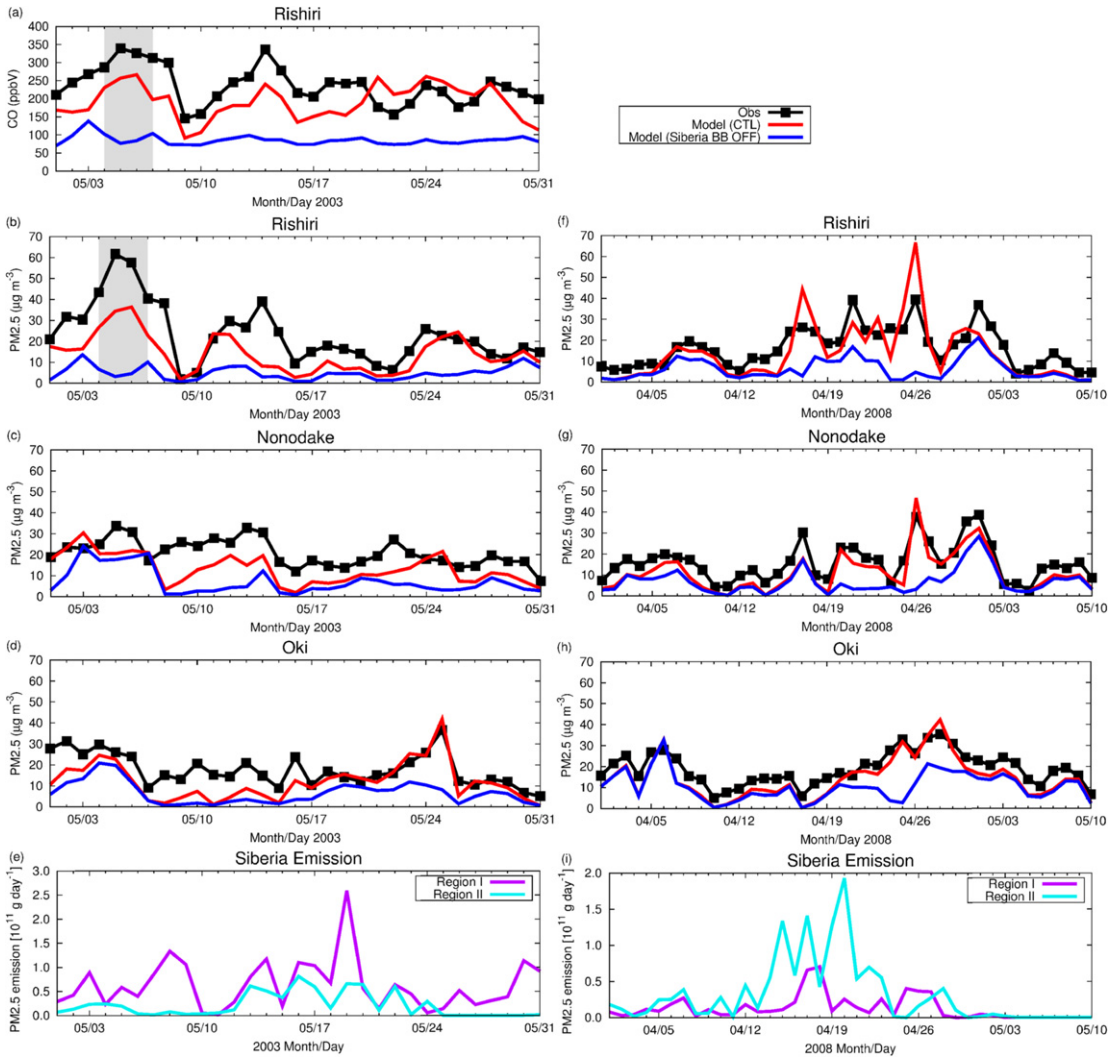

Figure 3. Temporal variations of observed and simulated daily mean $PM_{2.5}$ mass concentrations and CO mixing ratio at Rishiri, Nonodake, and Oki in May 2003 (a)–(d) and April 2008 (f)–(h). Observations are indicated by black lines. Model results of the control simulation and sensitivity run without Siberian biomass burning emissions are shown by red and blue lines, respectively. The gray areas denote the high event due to Siberian biomass burning on 4–6 May 2003. Daily biomass burning emissions of primary $PM_{2.5}$ from regions I (50°N–55°N, 105°E–120°E) and II (48°N–55°N, 122°E–135°E) are also shown in (e) and (i).

capability of models to predict $PM_{2.5}$ due to Siberian wildfires, in particular, emissions. Figure 5 shows observed monthly mean $PM_{2.5}$ concentrations and simulated $PM_{2.5}$ compositions at Rishiri in May 2003 and April 2008. In May 2003, the model largely underestimated $PM_{2.5}$ concentrations compared with observations. Some recent studies have reported that GFEDv3.1 underestimates emissions from wildfires in Siberia (e.g. Dolman *et al* 2012, Konovalov *et al* 2014). The uncertainties in the emission inventories could lead to negative biases for the days during which the influence of Siberian biomass burning was large (figure 3(b)), as mentioned above. It should be noted that $PM_{2.5}$ originating from the sources other than biomass burning in Siberia would also contribute to the model biases because the model tended to underestimate observations for the periods when the influence of Siberian wildfires was small (Ikeda *et al* 2014). By contrast, the model reproduced the concentration level reasonably well in 2008, in particular, for high $PM_{2.5}$ events due to Siberian biomass burning (figure 3(f)).

Although the reasons why the model reproduced well for the burning events in 2008 but not in 2003 are not identified here, these results might highlight that the GFED inventory underestimated the emissions in 2003 but reasonably predicted the emissions in 2008 in terms of locations and periods of time. Another possible reason is the differences in transport times from source regions to the measurement site (~1–2 days), which leads to the differences in secondarily formed $PM_{2.5}$. The distance between the major fire areas in May 2003 (east of Lake Baikal) and the observation site (~2000 km) was about two times greater than that in April 2008 (along River Amur) (figure 2). Many studies have reported that models including CMAQv4.7.1 underestimate the mass concentrations of secondary organic aerosols (SOA) (Carlton *et al* 2010, Hallquist *et al* 2009). Thus, more SOA that

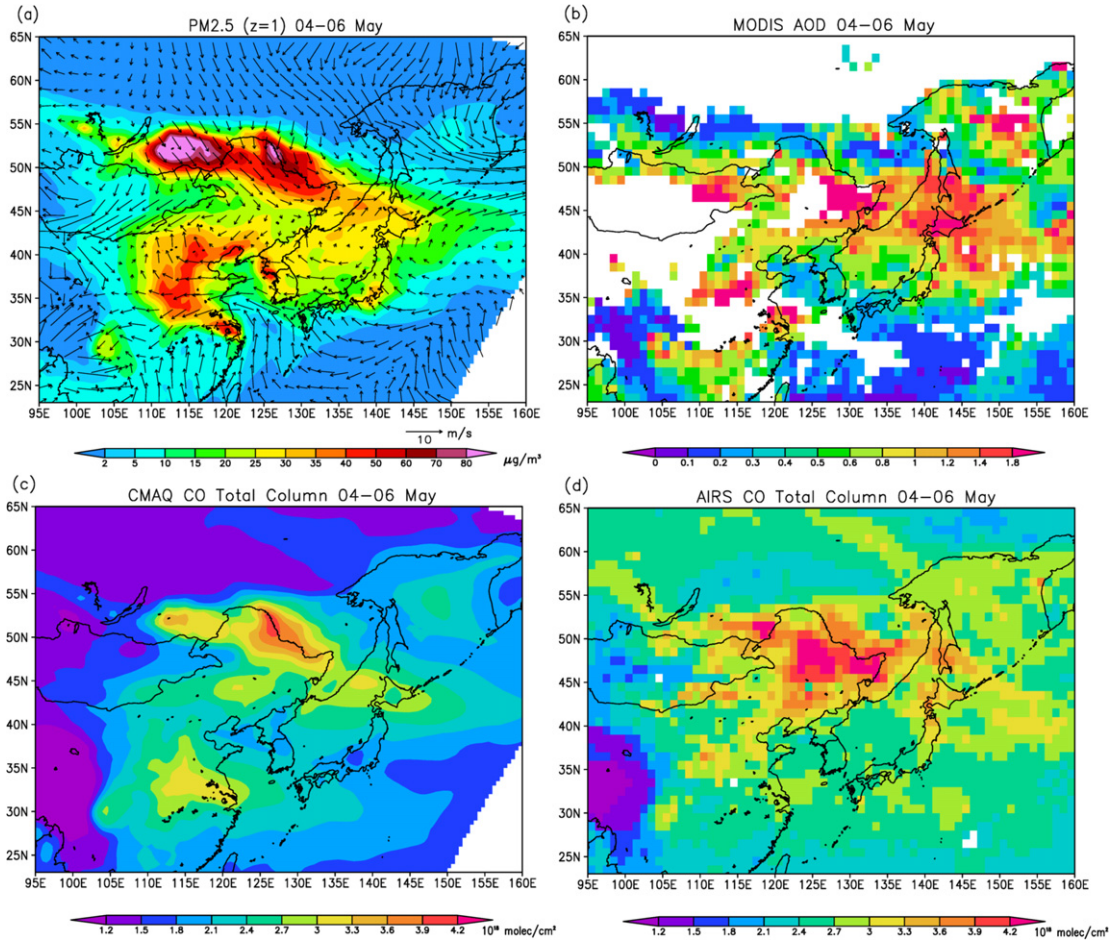

Figure 4. Horizontal distribution of surface $PM_{2.5}$ by model simulation (a), aerosol optical depth (AOD) from MODIS observations (b), CO total column by model (c), and CO total column from AIRS observations (d).

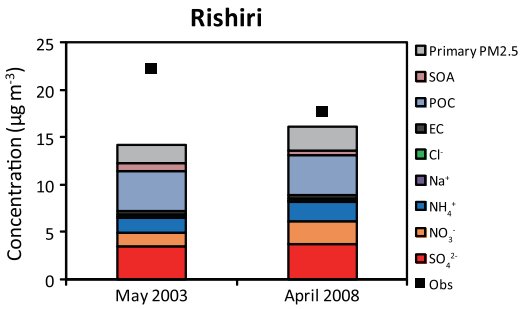

Figure 5. Monthly mean concentrations of $PM_{2.5}$ composition at Rishiri in the control simulations and observed total $PM_{2.5}$ mass concentrations in May 2003 and April 2008.

was not accurately represented by the model could be formed during a longer transport time, leading to a larger negative bias in May 2003. In addition, the aging process of POC that is not treated in the current model could be also more significant for May 2003, and thus the increase of noncarbon organic mass associated with POC would be larger than that in April 2008 (Simon and Bhave 2012).

Figure 6 shows the comparison of the observed $PM_{2.5}$ and AOD at the observation sites over Japan in

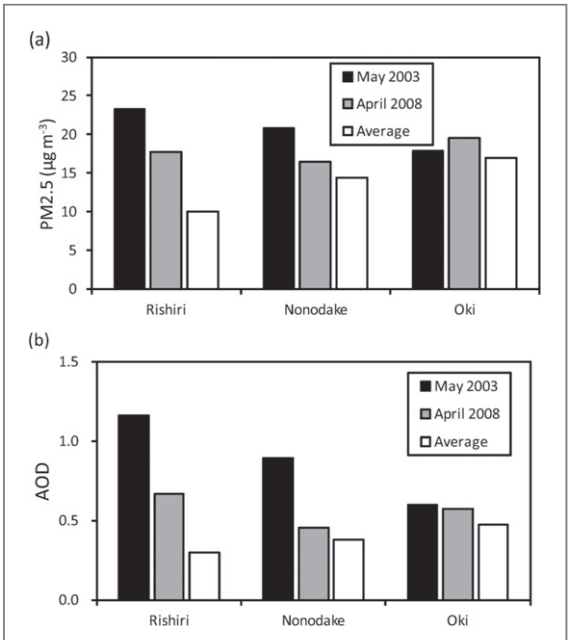

Figure 6. Monthly means of observed $PM_{2.5}$ concentrations (a) and aerosol optical depth (b) in May 2003, April 2008, and long-term average in the spring (April–May) from 2001 to 2012 at Rishiri, Nonodake, and Oki. The average values are without the intense fire years 2003 and 2008.

May 2003 and April 2008 with the long-term average in the spring (April–May) during 2001–2012. The monthly mean $PM_{2.5}$ concentrations for May 2003 and April 2008 were higher than the climatological values at every site. Especially at Rishiri, the observed result in May 2003 was more than two times larger than the long-term average of 10.1 $\mu g\,m^{-3}$. Ikeda *et al* (2015) reported that the annual mean $PM_{2.5}$ concentrations are generally higher in the western part of Japan and decrease toward the northeast of Japan by analyzing the observation data over Japan for the year 2010. It was also demonstrated that this significant geographic gradient of $PM_{2.5}$ concentrations was caused by the contribution of anthropogenic $PM_{2.5}$ transported from the Asian continent (Ikeda *et al* 2015). The spatial pattern observed in 2010 was consistent with the long-term averaged values in spring; $PM_{2.5}$ concentration was the lowest at Rishiri and highest at Oki. However, the results for May 2003 clearly showed that this trend in the horizontal distribution of $PM_{2.5}$ concentrations was reversed due to the influence of the intense Siberian wildfires during this period. The contribution of biomass burning in Siberia was also obvious in the AOD values over Japan, and the relationship between AOD in May 2003 and the long-term average was consistent with $PM_{2.5}$ mass concentrations. The AOD at Rishiri in May 2003 was three times higher than the climatological value, and much higher than the AOD values at other sites in Japan. Conversely, the AOD value of the long-term average was the smallest at Rishiri.

The annual average concentration at Rishiri in 2003 and 2008 were 19% and 11% larger than the long-term average, respectively (table 1), but the absolute concentrations of 9.8 and 9.1 $\mu g\,m^{-3}$ were still much lower than the Japanese air quality standard for annual mean $PM_{2.5}$ concentration (15 $\mu g\,m^{-3}$). Thus, this result suggests that influences of Siberian wildfires on Japan are not so large on an annual basis even for intense fire years. Ikeda *et al* (2015) estimated that the relative contributions from the total natural sources other than anthropogenic emissions (i.e. biomass burning, biogenic sources, and volcanoes) to the annual mean concentrations over Japan were small (1–6%) in 2010. Since the amount of primary $PM_{2.5}$ from Siberian biomass burning for the year 2010 (2.0 Tg $year^{-1}$) is nearly equal to the long-term average (2.2 Tg $year^{-1}$) (table 1), this result suggests that the contributions from Siberian wildfires to the annual average in Japan are negligible for the years without intense fires. Thus, the increases from the long-term average in 2003 and 2008 (i.e. 19% and 11%) correspond approximately to the relative contributions from Siberian biomass burning to the annual mean $PM_{2.5}$ concentrations in these intense fire years.

4. Conclusions

We investigated the influence of Siberian biomass burning on $PM_{2.5}$ pollution with a focus on air quality standard in Japan during 2001–2012. Several enhancements resulting in $PM_{2.5}$ concentrations exceeding Japan's air quality standard for daily mean value (35 $\mu g\,m^{-3}$) were observed at Rishiri Island in northern Japan in the spring of 2003 and 2008, when intense wildfires occurred in Siberia. AOD from MODIS and CO total column observed by AIRS showed that aerosols and CO originating from Siberian wildfires were concurrently transported from the source regions toward Japan, as observed at the surface of Rishiri. The model simulations with and without biomass burning emissions in Siberia also demonstrated that the elevated concentrations of $PM_{2.5}$ during the events were attributed mostly to Siberian biomass burning. Thus, it is suggested that the contribution from Siberian wildfires had a critical impact on $PM_{2.5}$ pollution resulting in daily mean concentrations over 35 $\mu g\,m^{-3}$. In other years except 2003 and 2008, the exceedance of the air quality standard due to Siberian wildfires was not observed. Although Siberian biomass burning does not affect the air quality standard of $PM_{2.5}$ for the years without intense wildfires, it causes exceedance of the air quality standard level if intense fires occur. The influence of Siberian biomass burning on $PM_{2.5}$ mass concentrations should be carefully monitored since fire activities in boreal forests are predicted to increase in future warming climate (Stocks *et al* 1998, Malevsky-Malevich *et al* 2008, Tchebakova *et al* 2009).

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