

1 **Application of a global nonhydrostatic model with a**
2 **stretched-grid system to regional aerosol simulations**
3 **around Japan**

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23 **Abstract**

24 An aerosol-coupled global nonhydrostatic model with a stretched-grid system has been
25 developed. Circulations over the global and target domains are simulated with a single
26 model, which includes fine meshes covering the target region to calculate meso-scale
27 circulations. The stretched global model involves lower computational costs to simulate
28 atmospheric aerosols with fine horizontal resolutions compared with a global uniform
29 nonhydrostatic model, whereas it may require higher computational costs compared
30 with the general regional models, because the stretched-grid system calculates inside
31 and outside the target domain. As opposed to general regional models, the
32 stretched-grid system does require neither a nesting technique nor lateral boundary
33 conditions. In this study, we developed a new-type regional model for the simulation of
34 aerosols over Japan, especially in the Kanto areas surrounding Tokyo, with a maximum
35 horizontal resolution of approximately 10 km. This model usually reproduces temporal
36 variations and their averages of the observed weather around Japan. This model
37 generally reproduces monthly mean distributions of the observed sulfate and SO₂ over
38 East Asia, with the high correlations ($R > 0.6$), but the underestimation of the simulated
39 concentrations by 40% (sulfate) and 50% (SO₂). Their underestimation of the simulated
40 sulfate and SO₂ concentrations over East Asia are strongly affected by their
41 underestimation in China and possibly by the uncertainty of the simulated precipitation
42 around Japan. In the Kanto area, this model succeeds in simulating the wind patterns
43 and the diurnal transitions around the center of the Kanto area, although it is inadequate
44 to simulate the wind patterns and the diurnal transitions at some sites located at the edge

45 of the Kanto area and surrounded on three sides by mountains, e.g., Maebashi, mainly
46 due to the insufficient horizontal resolution. This model also generally reproduces both
47 diurnal and synoptic variations of the observed and/or a regional aerosol-transport
48 model, WRF-CMAQ, simulated EC, sulfate, and SO₂ concentrations in the Kanto area,
49 especially with their high correlation ($R>0.5$) at Komae/Tokyo. Although the aerosol
50 module used in this study is relatively simplified compared to the general regional
51 aerosol models, this study reveals that our proposed model with the stretched-grid
52 system can be applicable for the regional aerosol simulation.

53 **1 Introduction**

54 Aerosols can greatly affect regional air quality and contribute to global climate change
55 (Forster et al., 2007). Recently, transboundary aerosol pollution, whereby regions
56 beyond a given country's borders are affected by the aerosols generated in that country,
57 has been of increasing concern (Ramanathan et al., 2008; Yu et al., 2012). The ongoing
58 rapid economic growth in developing countries has the potential to exacerbate this issue
59 (UNEP and WMO, 2011). Air pollution generated by aerosols is a critical public health
60 issue due to the deleterious effects of these particles on human health (Dockery et al.,
61 1993; Pope et al., 2009). Aerosols, which scatter and absorb solar radiation and act as
62 cloud condensation nuclei, can directly and indirectly change the Earth's radiation
63 budget. The majority of aerosols are emitted from localized areas, which are referred to
64 as hotspots, such as megacities and biomass-burning regions, and are spread throughout
65 the world via atmospheric transport (e.g., Ramanathan et al., 2008). Therefore, global
66 aerosol-transport models should consider the important regional-scale characteristics of
67 aerosol hotspots to reliably estimate their impacts on air quality and climate change.

68 Most existing global aerosol-transport models do not address the spatial variability of
69 aerosols in the vicinity of hotspots due to their coarse horizontal resolution of 100–300
70 km (Kinne et al., 2006; Textor et al., 2006). In addition, global aerosol-transport models
71 with coarse resolutions frequently adopt a spectral transform method with a hydrostatic
72 approximation to effectively calculate atmospheric dynamics. This spectral transform
73 method is less effective than the grid-point method (Stuhne and Peltier, 1996; Taylor et

74 al., 1997; Randall et al., 2000) for high horizontal resolutions (Tomita et al., 2008).
75 Models that employ the grid-point method flexibly define grid points to enable an
76 adaptive focus on study regions. Thus, global models based on the grid-point method
77 seem most appropriate for use in simulating aerosol transport from hotspots to outflow
78 regions.

79 For this purpose, we utilized the global Nonhydrostatic Icosahedral Atmospheric Model
80 (NICAM) developed by Tomita and Satoh (2004) and Satoh et al. (2008). NICAM has
81 been employed for the global simulation of atmospheric processes with high-resolution
82 grid spacing, whose size is comparable to the typical deep convective cloud scale.
83 Miura et al. (2007) performed a one-week computation with a horizontal resolution of
84 3.5 km using the Earth Simulator at the Japan Agency for Marine-Earth Science and
85 Technology (JAMSTEC) to successfully simulate a Madden-Julian Oscillation (MJO)
86 event. Suzuki et al. (2008) implemented an aerosol transport model named the Spectral
87 Radiation-Transport Model for Aerosol Species (SPRINTARS; Takemura et al., 2005)
88 in NICAM (we refer to this aerosol-coupled model as NICAM-SPRINTARS) and
89 performed a one-week simulation with a horizontal resolution of 7 km using the Earth
90 Simulator. Although these global, highly resolved calculations are promising with
91 regard to long-term climate simulations for decades, their requirement of vast computer
92 resources substantially limits their use in short-duration and/or case-specific simulations
93 due to the current limitations of computational resources. To overcome this limitation,
94 we adopt a compromise approach based on a new grid transformation named the

95 stretched grid system, which was developed and implemented in NICAM by Tomita
96 (2008a) for computationally effective simulations in the target region (see, also, Satoh
97 et al. 2010). We applied this approach to NICAM-SPRINTARS, which we named
98 Stretch-NICAM-SPRINTARS, to calculate aerosol transport processes with high
99 horizontal resolutions over aerosol source regions.

100 In this study, we focused on Japan, especially the Kanto region surrounding Tokyo
101 (Figure 1), because the Kanto region living more than 30 million people is one of the
102 largest megacities in the world. In Japan, a monitoring system for the air pollution, e.g.,
103 PM_{2.5} (aerosol particles with diameters less than 2.5 μm) and SO₂, has been operated
104 by the Japanese government. Inorganic ions, mainly sulfate, have been measured over
105 Japan and other Asian countries under EANET (Acid Deposition Monitoring Network
106 in East Asia; <http://www.eanet.asia/index.html>). Measurements of carbonaceous
107 aerosols were limited, with the exception of intensive measurements (Fine Aerosol
108 Measurement and Modeling in Kanto Area, FAMIKA) in the Kanto region during
109 summer 2007 (Hasegawa et al., 2008; Fushimi et al., 2011). For the model evaluation
110 using these measurements, we simulated aerosol spatial distributions during August
111 2007 using Stretch-NICAM-SPRINTARS with a horizontal resolution of approximately
112 10 km over the Kanto region. Because the model framework of
113 Stretch-NICAM-SPRINTARS is identical to that of globally uniform grid simulation
114 (we named it Global-NICAM-SPRINTARS), with the exception of the grid
115 configuration, and involves lower computational costs than global simulations, the

116 investigation of the model performance of Stretch-NICAM-SPRINTARS can be simply
117 and effectively extended to improve the original NICAM-SPRINTARS with globally
118 uniform high resolution for near-future simulations. To evaluate aerosol simulations
119 with the stretched-grid system, in this study we also conducted
120 Global-NICAM-SPRINTARS, but with relatively low resolution (approximately 100
121 km) due to the limited computational resources. The model intra-comparison approach,
122 with the exception of the grid system and the spatial resolution, is very meaningful to
123 investigate impacts of the stretched-grid system on the aerosol simulations. In addition,
124 Stretch-NICAM-SPRINTARS can be a new-type model that is also applicable for a
125 regional simulation of aerosols, because it focuses on a specific regional domain
126 without require a nesting technique nor boundary conditions, unlike general regional
127 models.

128 For the model evaluation in the target Japan, we mainly focused on a representative
129 primary aerosol, i.e., elemental carbon (EC), and a representative secondary aerosol, i.e.,
130 sulfate. EC is directly emitted from anthropogenic combustion processes, and is a good
131 indicator to monitor the transport pattern. The global and regional modelings for sulfate,
132 which is formed from SO_2 in the atmosphere, are more deeply understood compared to
133 modelings for the other secondary aerosols such as nitrate and organic aerosols (e.g.,
134 Barrie et al., 2001; Holloway et al., 2008; Hallquist et al., 2009; Morino et al., 2010a,
135 2010b). In addition, sulfate is the largest contributor to the total secondary inorganic
136 aerosols (e.g., Zhang et al., 2007), and the sulfate mass concentrations are larger than

137 that the nitrate ones in August 2007 over the Kanto area (Morino et al., 2010c).
138 Originally, these basic components (EC and sulfate) are suitable for the evaluation in
139 this study, primarily because the stretched-grid system was applied to the simulations of
140 atmospheric pollutants over the land in the mid-latitude band for the first time and
141 secondly because the original SPRINTARS is more simplified compared to
142 conventional regional aerosol models.

143 This paper is organized as follows: the model framework of NICAM and SPRINTARS
144 and the experimental design are described in Section 2. We show two model results; (1)
145 Stretch-NICAM-SPRINTARS with glevel-6, in which “glevel” is the number of
146 divisions of an icosahedron used to construct the horizontal grid, (hereafter referred to
147 as the “NICAM-g6str” model) and (2) Global-NICAM-SPRINTARS with glevel-6
148 (hereafter referred to as the “NICAM-g6” model). In Section 3, the model results are
149 validated using in-situ measurements in terms of meteorological fields including
150 precipitation and aerosol species, especially EC, sulfate and SO₂. For the model
151 evaluation of chemical species, we also made use of results in a regional aerosol model,
152 the Community Multiscale Air Quality (CMAQ) driven by the Weather Research and
153 Forecasting (WRF) model named WRF-CMAQ, shown by Shimadera et al. (2013). We
154 also present the validation of total aerosol amounts, i.e., PM_{2.5}, and aerosol optical
155 product, i.e., extinction for spherical aerosols. Finally, the conclusions are summarized
156 in Section 4.

157

158 **2 Model description**

159 **2.1 Nonhydrostatic Icosahedral Atmospheric Model (NICAM)**

160 NICAM, which employs an icosahedral grid-point method with a nonhydrostatic
161 equation system (Tomita and Satoh, 2004; Satoh et al., 2008, 2014), is run with a
162 maximum horizontal resolution of 3.5 km (Tomita et al. 2005; Miura et al., 2007) and
163 can be applied to a transport model of aerosols and gases as a conventional atmospheric
164 general circulation model (Suzuki et al., 2008; Niwa et al., 2011; Dai et al., 2014a,
165 2014b; Goto, 2014). NICAM can also be employed for regional-scale simulations by
166 adopting a stretched-grid system (Tomita, 2008a; Satoh et al., 2010). The stretched
167 icosahedral grid was developed from a general grid transformation method, i.e., the
168 Schmidt transformation method, for a horizontal grid system on a sphere. In the
169 Schmidt transformation, the grid interval on a sphere lacks uniformity with a finer
170 horizontal resolution close to the center of the target region. Tomita (2008a) showed
171 that the Schmidt transformation minimizes potential errors involving the isotropy and
172 homogeneity of the target region. The stretched-grid system can solve the main
173 problems associated with commonly used regional models, which occur from artificial
174 perturbations near boundary areas in cases where meteorological and aerosol fields are
175 prescribed. In addition, the computational cost of the stretched-grid system is
176 substantially lower than that of a global calculation under the same horizontal resolution
177 in the target region. For example, when the globally uniform grid with a maximum
178 horizontal resolution of 10 km is applied to the global simulation, the minimum

179 required theoretical computational cost is 64-256 times higher than the cost of the
180 stretched-grid system in this study. Compared to conventional regional models, the
181 computational cost may increase because the stretched-grid system requires the
182 calculation outside the target domain. Furthermore, the model framework of the
183 stretched global model is identical to that of the uniformed global model without special
184 modifications, whereas the model framework of regional models is usually different
185 from that of global models. These advantages can facilitate additional developments for
186 global simulations by testing a new scheme with minimal computational cost.
187 Compared with general regional models, the stretched-grid system is more suitable for
188 the future study, which aimed to extend its use to the global uniform high-resolution
189 NICAM-SPRINTARS.

190 In this study, we adopt the stretched-grid system to focus on the Kanto region, including
191 Tokyo, using glevel-6 resolution and the stretched ratio of 100 (we call it
192 NICAM-g6str), which is the ratio of the largest horizontal grid spacing located on the
193 opposite side of the earth from Tokyo to the smallest horizontal grid spacing near
194 Tokyo. As a result, a minimum horizontal resolution of 11 km around the center
195 (140.00°E, 35.00°N) was used. NICAM implements comprehensive physical processes
196 of radiation, boundary layer and cloud microphysics. The radiation transfer model is
197 implemented in NICAM with the k-distribution radiation scheme MSTRN, which
198 incorporates scattering, absorption and emissivity by aerosol and cloud particles as well
199 as absorption by gaseous compounds (Nakajima et al., 2000; Sekiguchi and Nakajima,

200 2008). The vertical turbulent scheme comprises the level 2 scheme of turbulence closure
201 by Mellor and Yamada (1974), Nakanishi and Niino (2004, 2009) and Noda et al.
202 (2009). The cloud microphysics consist of the six-class single-moment bulk scheme
203 (water vapor, cloud water, rain, cloud ice, snowflakes and graupel) (Tomita, 2008b).
204 Based on our experience in previous studies, we did not employ cumulus
205 parameterization in this study (e.g., Tomita et al., 2005; Sato et al., 2009; Nasuno, 2013).
206 The topography used in this study is based on GTOPO30 (the horizontal resolution is 30
207 arc seconds, that is approximately 1 km) courtesy of the U.S. Geological Survey. The
208 vertical coordinates system adopts Lorenz grid and z^* (terrain-following) coordinates
209 with the 40 layers of z-levels and model top of 40 km height (Satoh et al., 2008). The
210 timestep was set to 20 seconds.

211

212 **2.2 SPRINTARS**

213 Based on the approach of Suzuki et al. (2008), the three-dimensional aerosol-transport
214 model—Spectral Radiation-Transport Model for Aerosol Species (SPRINTARS;
215 Takemura et al., 2000, 2002, 2005; Goto et al. 2011a,b,c)—was coupled to NICAM in
216 this study. The SPRINTARS model calculates the mass mixing ratios of the primary
217 tropospheric aerosols, i.e., carbonaceous aerosol (EC and OC, organic carbon), sulfate,
218 soil dust, sea salt and the precursor gases of sulfate, namely, SO_2 and dimethylsulfide
219 (DMS). The aerosol module considers the following processes; emission, advection,
220 diffusion, sulfur chemistry, wet deposition and dry deposition, including gravitational
221 settling. For carbonaceous aerosols, the 50% mass of EC from fossil fuel sources is

222 composed of externally mixed particles, whereas other carbonaceous particles are
223 emitted and treated as internal mixtures of EC and OC (EC-OC internal mixture).
224 Biogenic secondary organic aerosols (SOAs) from terpenes are treated but are greatly
225 simplified by multiplying a conversion factor to the terpenes emission (Takemura,
226 2012). In addition, anthropogenic SOAs from toluene and xylene are disregarded in this
227 study. The bulk mass concentrations of EC, OC, and sulfate are calculated by
228 single-modal approach, which means that the SPRINTARS model does not explicitly
229 treat aerosol dynamic processes such as coagulation and condensation. The particle size
230 distribution of the dry particles are prescribed in a logarithmic normal size distribution
231 with dry mode radii of 18, 100, 80 and 69.5 nm, for pure EC, EC-OC internal mixture,
232 biogenic SOA and externally mixed sulfate, respectively (Goto et al., 2011a). The
233 hygroscopicities, densities and refractive indices for the aerosols are set to the same
234 values used by Takemura et al. (2002) and Goto et al. (2011a). The combinations of the
235 pre-calculated cross-sections of the extinction and simulated mixing ratios for each
236 aerosol species provide the simulated aerosol extinction coefficient for each timestep of
237 the model (Takemura et al., 2002). The sulfur chemistry in SPRINTARS considers only
238 three chemical reactions to form sulfate through gas-phase oxidation of SO_2 by
239 hydroxyl radical (OH) and aqueous-phase oxidation by ozone and hydrogen peroxide.
240 The large part of SO_2 are emitted from fossil fuel combustion, biomass burning, and
241 volcano eruption, whereas some of SO_2 are formed from the oxidation of DMS, which
242 is emitted naturally from marine phytoplanktons. The numerical solution in the
243 oxidations adopts an approximation in a quasi first-order reaction using the same

244 integrated time resolution as that of the dynamic core. The pH value in the
245 aqueous-phase is fixed at 5.6, because the SPRINTARS model treats limited ions in the
246 aqueous-phase (e.g., Takemura et al., 2000). The oxidant distributions (OH, ozone and
247 hydrogen peroxide) were offline provided by a chemical transport model. The
248 atmospheric removal of aerosols in SPRINTARS includes wet (due to rainout and
249 washout) and dry (due to turbulence and gravity) deposition processes, whereas those of
250 SO₂ only include rainout and dry deposition by turbulence. In the cloudy grid, the mass
251 fractions of sulfate out of the cloud droplets to the mass of sulfate in the grid were fixed
252 at 0.5, whereas the fractions for SO₂ were determined by Henry's law (Takemura et al.,
253 2002). As for pure EC, EC-OC internal mixture, and biogenic SOA, the mass fractions
254 were fixed at 0.1, 0.3, and 0.3, respectively. Because the SPRINTARS model does not
255 predict the mass mixing ratio of the chemical tracers inside the clouds, it assumes that
256 the tracers inside the clouds are evaporated from the clouds at one timestep. In this
257 study, the particle mass concentrations for diameters less than 2.5 μm (defined as
258 PM_{2.5}) are calculated by summing EC, organic matter by multiplying OC by 1.6
259 (Turpin and Lim, 2001), sulfate and ammonium aerosols. Because this model cannot
260 directly predict ammonium compounds, it is assumed that all sulfate is the form of
261 ammonium sulfate, so that their concentration was estimated by multiplying the mass
262 concentration of sulfate by 0.27, which is the molar ratio of ammonium ion to
263 ammonium sulfate. The nitrate in this study is disregarded, primarily because the main
264 objective in this study is modeling of sulfate as a representative secondary aerosols and
265 secondly because the nitrate mass concentrations are lower than the sulfate ones with

266 the target of August 2007 in Japan (Morino et al., 2010c).

267

268 **2.3 Design of the experiments**

269 The target period comprises one month in August 2007, in which an intensive
270 measurement of aerosol chemical species was conducted under Project FAMIKA
271 (Hasegawa et al., 2008; Fushimi et al., 2011). The six-hour meteorological fields (wind
272 and temperature) were nudged above a height of 2 km using NCEP-FNL reanalysis data
273 (<http://rda.ucar.edu/datasets/ds083.2/>). The one-day sea surface temperature was also
274 nudged using the NCEP-FNL data. The initial conditions were prescribed by the
275 NCEP-FNL data for the meteorological fields and the one and a half months spinup
276 results of the Stretch-NICAM-SPRINTARS model for the aerosol fields, respectively.

277 The emission inventories of anthropogenic EC, OC and SO₂ in this experiment were
278 prepared by EAGrid2000 with a horizontal resolution of 1 km over Japan (Kannari et al.,
279 2007), REAS version 2 with a horizontal resolution of 0.25° over Asia (Kurokawa et al.,
280 2013) and the AeroCom inventory with a horizontal resolution of 1° over other areas of
281 the world (Diehl et al., 2012). Because EAGrid2000 does not explicitly estimate EC and
282 OC inventories, we estimated the inventories to be consistent with those from previous
283 studies (Morino et al., 2010a,b; Chatani et al., 2011) by modifying the PM_{2.5} inventory
284 of EAGrid2000 using scaling factors of EC/PM_{2.5} and OC/PM_{2.5} based on sources.
285 These inventories of anthropogenic EC and SO₂ in 2007 are described in Figure 2. The
286 emissions of SO₂ from volcanoes in Japan, such as Miyakejima and Sakura-jima, were

287 obtained from statistical reports (<http://www.seisvol.kishou.go.jp/tokyo/volcano.html>)
288 by the Japan Meteorological Agency (JMA). In this study, the distributions of three
289 hourly averaged monthly oxidants (OH, ozone and hydrogen peroxide) were derived
290 from a global chemical transport model (CHASER) coupled to the Model for
291 Interdisciplinary Research on Climate (MIROC), named MIROC-CHASER, with the
292 spatial resolution of 2.8° by 2.8° (Sudo et al., 2002).

293 To evaluate model performances in the stretched-grid system, we also simulated
294 NICAM-SPRINTARS with the globally uniformed grid simulation in glevel-6
295 resolution (the horizontal resolution is set to 110 km and we call it NICAM-g6).
296 Global-NICAM-SPRINTARS with relatively low resolution has been applied to aerosol
297 simulations and well compared with in-situ measurements and satellite remote sensing
298 (Dai et al., 2014a; Goto, 2014). Apart from the NICAM-g6str simulation, in the
299 NICAM-g6 simulation, the cloud physics apply both the prognostic
300 Arakawa-Schubert-type cumulus convection scheme (Arakawa and Schubert, 1974) and
301 the diagnostic large-scale clouds described by Le Treut and Li (1991). The large-scale
302 cloud module is based on single moment bulk scheme for cloud mixing ratio. The
303 precipitation rate is parameterized by Berry (1967). Except for the grid system and the
304 horizontal resolution (which determines the module of the cloud physics),
305 Global-NICAM-SPRINTARS was identical to Stretch-NICAM-SPRINTARS.
306 Therefore, the comparison between NICAM-g6str and NICAM-g6 led to clarify
307 impacts of the horizontal resolution on the aerosol distribution.

308

309 **2.4 Observation**

310 In this study, we focused on the aerosol chemical component of EC as the primary
311 particle and sulfate as the secondary particle. To evaluate the model results over the
312 Kanto region, we used observations of the surface mass concentrations of EC and
313 sulfate in four cities under Project FAMIKA: Maebashi/Gunma (139.10°E, 36.40°N),
314 Kisai/Saitama (139.56°E, 36.09°N), Komae/Tokyo (139.58°E, 35.64°N) and
315 Tsukuba/Ibaraki (140.12°E, 36.05°N). The EC particles in PM_{2.5} were collected every
316 six hours with quartz fiber filters and analyzed with the thermal/optical method
317 according to the IMPROVE protocol (Chow et al., 2001). The sulfate particles in PM_{2.5}
318 were also collected every six hours with Teflon filters and analyzed by ion
319 chromatography. In addition to the limited FAMIKA dataset, we utilized measurements
320 taken by the EANET (Acid Deposition Monitoring Network in East Asia;
321 <http://www.eanet.asia/index.html>) and the 4th national survey report of acid rain over
322 Japan in fiscal year 2007
323 (http://tenbou.nies.go.jp/science/institute/region/journal/JELA_3403041_2009.pdf) to
324 assess the monthly mean concentrations of sulfate and SO₂ at Japanese and Korean sites.
325 We also obtained Chinese measurements by *Zhang et al.* [2012], as part of the Chinese
326 Meteorological Administration Atmosphere Watch Network (CAWNET). To validate
327 the concentration of SO₂ for the Kanto region, we accessed monitoring stations operated
328 by Japanese and local governments.

329 In the validation of the meteorological fields simulated by NICAM-g6str and
330 NICAM-g6, we used meteorological fields (wind and temperature) reanalyzed by
331 NCEP-FNL over East Asia. In the Kanto region, we obtained measurements for the
332 meteorological parameters (temperature, relative humidity (RH) and wind) at or near
333 the 7 sites of Project FAMIKA and additional cities: Tsuchiura/Ibaraki (140.20°E,
334 36.07°N), which is the city nearest to Tsukuba; Yokohama/Kanagawa (139.64°E,
335 35.45°N); Chiba/Chiba (140.12°E, 35.62°N); Adachi/Tokyo (139.82°E, 35.77°N); and
336 Machida/Tokyo (139.43°E, 35.53°N), which is the city nearest to Komae, as shown in
337 Figure 1(b). For precipitation, we used a measurement taken by the Automated
338 Meteorological Data Acquisition System (AMeDAS) at 21 sites over Japan including
339 the following 10 Kanto's sites: Yokohama; Chiba; Tsukuba; Tokyo, which is near
340 Adachi; Maebashi; Huchu, which is near Machida; Konosu, which is near Kisai; Abiko
341 (140.11°E, 35.60°N); Saitama (139.59°E, 35.88°N); and Nerima (139.59°E, 35.74°N)
342 (Figure 1). To evaluate the spatial patterns of the precipitation obtained by
343 NICAM-g6str and NICAM-g6, we used the quantities of the monthly mean
344 precipitation around Japan that were derived from the Global Satellite Mapping of
345 Precipitation (GSMaP; Okamoto et al., 2005; Kubota et al., 2007; Aonashi et al., 2009;
346 Ushio et al., 2009) and the Meso Scale Model (MSM) developed by the JMA for rain
347 forecast (Saito et al., 2006). The results by MSM are generally higher accurate than
348 those in GSMaP, although the covering area in MSM is limited around Japan.

349 To evaluate the quantities of the total aerosol amounts, such as PM2.5, we compared the

350 simulated PM_{2.5} concentrations with the observations at the 18 sites including the
351 FAMIKA sites and other monitoring stations operated by the Japanese and local
352 governments (Figure 1). The PM_{2.5} concentrations were continuously observed using
353 tapered element oscillating microbalance (TEOM) with Series 1400a Ambient
354 Particulate Monitor. The instruments are controlled under the temperature of 50 °C, to
355 minimize the influence of change in the ambient temperature and RH. However, it
356 includes large uncertain due to the difficulty in completely eliminate the water content
357 attached to aerosols and lacks of the calibration of the instrument in some of sites.
358 Nevertheless, the observed PM_{2.5} concentrations with hourly time resolution were still
359 useful to validate the model results.

360 In Tsukuba and Chiba, light detection and ranging (LIDAR) measurements operated by
361 the National Institute for Environmental Studies (NIES) of Japan were also available
362 (Sugimoto et al., 2003; Shimizu et al., 2004). The LIDAR unit measured vertical
363 profiles of the backscattering intensity at 532 and 1064 nm and the depolarization ratio
364 at 532 nm. The backscattering intensity was converted to the extinction coefficient, and
365 the depolarization ratio distinguished the extinction between spherical and non-spherical
366 particles. In this study, we only used vertical profiles of the extinction for spherical
367 particles. A detailed algorithm was provided by Sugimoto et al. (2003) and Shimizu et
368 al. (2004).

369

370 **3 Validation of Stretch-NICAM-SPRINTARS**

371 **3.1 Meteorological fields**

372 So far, the stretched-grid system was mainly applied to the simulations of tropical
373 cyclones or tropical convective clouds with small domain over oceans for the short-term
374 period (less than several days) (e.g., Satoh et al., 2010; Arakane et al., 2014). In this
375 study, we focused on the air pollution around Japan (for the longer period). Therefore,
376 we first focused on the general circulation of the basic meteorological fields over the
377 large domain, which can affect the air pollution over Japan. Figure 3 shows temperature
378 and winds near the surface and the model height of approximately 5 km over Asia
379 region (100°E-170°E, 10°N-50°N). In August, North Pacific High (or Ogasawasa High)
380 mainly brings clear weather around Japan. A frequency of the precipitation is usually
381 limited, but a total amount of the monthly mean precipitation is not small, because of
382 typhoons and shower rain. In the focusing region, the general meteorological fields
383 simulated by NICAM-g6str and NICAM-g6 are comparable to those obtained by
384 NCEP-FNL. The absolute biases in the temperature between NICAM-g6str and
385 NCEP-FNL or between NICAM-g6 and NCEP-FNL are within 1.5 °C at the surface
386 and the height of 5 km. Around the Japanese Alps, however, the
387 NICAM-g6str-simulated temperature is lower than the NCEP-FNL-estimated one by at
388 most 2.5 °C, because of the differences in the resolved topography due to the different
389 spatial resolution between NICAM-g6str and NCEP-FNL. As for wind, western winds
390 over the northeastern part of Japan in both NICAM-g6str and NICAM-g6 are stronger
391 compared to those in NCEP-FNL. With the exception of this bias, the performances of

392 both NICAM-g6str and NICAM-g6 are good. Therefore, it is concluded that the
393 stretched-grid systems does not affect the general circulations under the nudging
394 technique in this study.

395 To evaluate the model performances of the six-hourly mean concentrations of aerosol
396 chemical species and SO₂ over the main target region, i.e., Kanto area, we used the
397 six-hourly instant observations of temperature, RH, wind and precipitation at each
398 station over the Kanto area shown in Figure 1. The results and summary are shown in
399 Figures 4 to 7 and Table 1. The NICAM-g6 results, especially in terms of diurnal
400 variations, tend to be far from the observations compared to the NICAM-g6str results,
401 because NICAM-g6, with the horizontal resolution of approximately 100 km, does not
402 fully resolve the topology over the Kanto area. Figure 4 illustrates the temporal
403 variations of temperature at a height of 2 m. The temporal variations in the
404 NICAM-g6str-simulated temperature are generally comparable to those in the observed
405 temperatures with root-mean-square-error (RMSE) values of less than 3°C, with the
406 exception of the results obtained for Maebashi and Machida. At these two sites, the
407 mean values of the NICAM-g6str-simulated temperatures are lower than those of the
408 observed temperatures by a maximum of 3.6°C. The correlation coefficients (R)
409 between NICAM-g6str and the observation range from 0.7–0.9, whereas the R between
410 NICAM-g6 and the observation range from 0.7–0.8, as shown in Table 1. Figure 5
411 shows the temporal variations in RH at a height of 2 m. The temporal variations in the
412 NICAM-g6str-simulated RH are similar to the observations, with the RMSEs in the

413 range of 10–15%. In contrast, the NICAM-g6-simulated RH is overestimated compared
414 to the observations, with the RMSEs in the range of 16–26%. The R values of RH
415 between the simulation (both NICAM-g6str and NICAM-g6) and observations are
416 approximately 0.6–0.8 (Table 1).

417 The temporal variations in the wind direction and speed simulated by NICAM-g6str are
418 compared with the observations in Figures 6 and 7. Near the southern part of the Kanto
419 area (Yokohama, Tsuchiura, Adachi and Machida), with the exception of Chiba, the
420 NICAM-g6str-simulated wind direction is generally comparable to the observations,
421 with a slight overestimation of the both NICAM-g6str and NICAM-g6 simulated wind
422 speed compared with the observations. At these four sites, the R and RMSE values in
423 NICAM-g6str range from approximately 0.5–0.7 and approximately 1.7–2.3 m/s,
424 respectively. In Chiba located near the ocean, the R value of wind speed between
425 NICAM-g6str and the observation is 0.41, whereas the NICAM-g6str-simulated wind
426 directions generally agree with the observations. Conversely, at Maebashi and Kisai, the
427 daily variations in the both NICAM-g6str and NICAM-g6 simulated wind directions
428 differ significantly from those in the observations, in which the southern winds and
429 northern winds frequently occur during the day and night, respectively, for example,
430 during August 5–12. At these two sites, the NICAM-g6-simulated wind direction and
431 speed is not closer to the observations compared to those obtained by NICAM-g6str.
432 The R value for wind speed between the NICAM-g6str and the observations at these
433 sites is estimated to be approximately 0.2. The observed southeasterly wind is long sea

434 breeze toward Maebashi Plateau surrounded on three sides by mountains around
435 Maebashi. The observed winds are caused by daytime meso-scale thermal lows
436 developed over the central Japan covering the Japanese Alps (Kuwagata and Sumioka,
437 1991). The Japanese Alps with the highest terrain in Japan can affect the local
438 meteorological fields even around 100-200 km away (Kitada et al., 1998). Therefore, it
439 suggests that the horizontal resolution in this study using NICAM-g6str (10 km over the
440 Kanto area) does not fully resolve the complex terrains of the Japanese Alps and the
441 Maebashi plateau. Therefore, it suggests that it is inadequate to simulate the wind
442 patterns and the diurnal transitions near high mountains around the Kanto area, whereas
443 it is adequate to simulate them around the center of the Kanto area.

444 Figures 8-10 show comparisons of NICAM-g6str and NICAM-g6 simulated
445 precipitation with the observations. Figure 8 compares the simulated precipitation with
446 the MSM and GSMaP derived results. During the early August 2007, mainly due to
447 passing of a typhoon over the western Japan, Okinawa, and Korea, the August mean
448 precipitation in the western Japan is larger than that in the eastern Japan, especially the
449 Kanto area. The monthly mean precipitation is estimated to be more than 200
450 mm/month over the western Japan, whereas that is estimated to be less than 50
451 mm/month over the eastern Japan. The horizontal patterns of the precipitation obtained
452 by NICAM-g6str in East China Sea, Sea of Japan near the Japan coast, and Korea are
453 closer to those derived from MSM and GSMaP than those obtained by NICAM-g6. In
454 the Kanto area, however, the NICAM-g6str-simulated precipitation with the range of

455 50-200 mm/month is overestimated compared to the MSM and GSMaP results. The
456 NICAM-g6-simulated precipitation over the Kanto area with the range of 100-200
457 mm/month is also much overestimated. In Figure 9 showing the temporal variations in
458 the amount of precipitation per day at 21 Japanese sites, the observed precipitation is
459 extremely limited during August 7-19 in the Kanto area. In other regions, the magnitude
460 of the precipitation is strong, although the precipitation is sporadic. In terms of the
461 frequency of the precipitation, the NICAM-g6str performance is better than the
462 NICAM-g6 one. Figure 10 illustrates the predictive value of daily precipitation, defined
463 as the ratio of the number of days where the model correctly predicts the weather (less
464 than 1 mm/day or more than 1 mm/day) to the number of the whole days. In the
465 NICAM-g6str results, the predictive values at most of sites over the Kanto area and four
466 sites over the non-Kanto area such as Nagoya and Osaka are calculated to be more than
467 85%. The predictive values obtained by NICAM-g6str are mostly higher than those
468 obtained by NICAM-g6. During the rainy days such as August 20, 22 and 23 over the
469 Kanto area, both NICAM-g6str and NICAM-g6 capture the precipitation, whereas
470 NICAM-g6str reproduces greater amounts of the precipitation and NICAM-g6
471 reproduces longer periods and larger areas compared to the observations. NICAM-g6str
472 does not always capture a sudden shower, as general meteorological models have
473 difficulties in predicting this type of precipitation system (e.g., Kawabata et al., 2011).
474 To increase the accuracy of such precipitation, more sophisticated cloud-microphysics
475 model, e.g., NICAM-NDW6 model proposed by Seiki and Nakajima (2014) based on
476 the double-moment bulk scheme with six water categories, may be required. In the

477 western Japan, during the rainy days, e.g., August 22-23, both NICAM-g6str and
478 NICAM-g6 usually capture large-scaled precipitation (Figure 9). Overall, NICAM-g6str
479 usually reproduces the observed weather in the target regions and periods, whereas
480 NICAM-g6 does not capture general feature such as the sporadic precipitation.

481

482 **3.2 Aerosol fields**

483 **3.2.1 Evaluation of chemical species**

484 Figures 11, 12, and 13 illustrates the temporal variations in the surface EC, sulfate, and
485 SO₂ concentrations at the four stations (Maebashi, Kisai, Komae and Tsukuba) in the
486 Kanto area using the simulations and the measurements. The simulations include
487 NICAM-g6str, NICAM-g6, and the Community Multiscale Air Quality (CMAQ) driven
488 by the Weather Research and Forecasting (WRF) model named WRF-CMAQ shown by
489 their Figures 5 and 6 of Shimadera et al. (2013). Shimadera et al. (2013) calculated the
490 WRF-CMAQ with a horizontal resolution of 5 km and an emission inventory that is
491 similar to that in the present study. Table 2 summarizes the statistical parameters for the
492 concentrations of EC, sulfate, and SO₂. The temporal variation and the average of EC
493 simulated by NICAM-g6str are better agreement with the observations obtained for
494 Komae than those simulated by NICAM-g6 (Figure 11(c)). However, the averages of
495 both NICAM-g6str and NICAM-g6 simulated EC concentrations at the other sites are
496 much underestimated compared to the observations (Table 2). For Tsukuba shown in
497 Figure 11(d), both the NICAM-g6str and NICAM-g6 simulated EC concentrations tend

498 to be underestimated compared with the observed concentrations, especially during the
499 daytime, even though the temporal variation of EC obtained by NICAM-g6str is closer
500 to the observed one compared to those obtained by NICAM-g6. At Maebashi and Kisai,
501 the temporal variation and the averages of EC obtained by NICAM-g6 are also
502 underestimated compared with the observations by a factor of three to five.
503 NICAM-g6str tends to have daily maximums of EC concentrations during the morning
504 time, whereas NICAM-g6 tends to have daily maximums during the nighttime. The
505 temporal variations of NICAM-g6str-simulated EC concentrations are generally
506 comparable to those by WRF-CMAQ shown in Figure 11 and their Figure 3 of Chatani
507 et al. (2014), with the exception of the results at Maebashi and Kisai where the EC
508 concentrations obtained by NICAM-g6str are smaller than those obtained by
509 WRF-CMAQ. At these sites, the difference in the EC concentrations between
510 NICAM-g6str and WRF-CMAQ is probably caused by the difference in the horizontal
511 resolution, which is most likely critical for properly simulating the air pollution
512 delivered by the meteorological wind fields from the center of the Kanto region (Kusaka
513 and Hayami, 2006). Table 2 also shows that the R obtained by NICAM-g6str at all sites
514 are high or moderate, with the exception of Maebashi, whereas those obtained by
515 NICAM-g6 and CMAQ are low. At most sites, the EC concentrations obtained by
516 WRF-CMAQ shown in Figure 11, and WRF-CMAQ illustrated by Morino et al.
517 (2010a,b) and Chatani et al. (2014), NICAM-g6str, and NICAM-g6 are also
518 underestimated compared to the observations with the larger values of RSME. The
519 underestimation of EC concentrations is investigated by sensitivity tests of EC emission

520 inventory in section 3.2.2.

521 At the same four sites, simulated sulfur components (sulfate and SO₂) are compared
522 with the observations in Figures 12 and 13. The observed SO₂ represents the ensemble
523 results of monitoring stations operated by Japanese and local governments around each
524 FAMIKA site. The mean differences in the sulfate mass concentrations between
525 NICAM-g6str and the observations are within approximately 10% at Maebashi and
526 Tsukuba, approximately -30% at Komae, and approximately +40% at Kisai. At all sites,
527 the temporal variations of the NICAM-g6str-simulated sulfate concentrations are
528 generally comparable to those obtained by the observations and WRF-CMAQ shown in
529 Figure 12 (i.e., their Figure 6 of Shimadera et al., 2013) and illustrated in their Figure 3
530 of Morino et al. (2010a), whereas the differences in the sulfate concentrations between
531 NICAM-g6str and the observations are somewhat greater on August 7 and 8 at
532 Maebashi where the performance of NICAM-g6str is relatively poor, mainly due to the
533 inadequate horizontal resolution to reproduce the observed meteorological fields, as
534 shown in section 3.1. The use of the prescribed distributions of three hourly averaged
535 monthly oxidants may partly cause the discrepancy of the hourly variations of the
536 sulfate between NICAM-g6str and the observations. The R obtained by all the models
537 (NICAM-g6str, NICAM-g6, and WRF-CMAQ) is acceptable at most sites, with the
538 exception of NICAM-g6str at Maebashi and WRF-CMAQ at Kisai. The RMSEs
539 obtained by all the models are smaller at Komae and Tsukuba than those at Maebashi
540 and Kisai. The six-hourly variations of the sulfate obtained by WRF-CMAQ are

541 sometimes missed by NICAM-g6str, partly due to the use of the prescribed oxidants.
542 Even though NICAM-g6 reproduces the synoptic cycle of the observed sulfate, it has
543 difficulties in simulating the diurnal cycle of the observed and NICAM-g6str-simulated
544 sulfate, as shown in the results of EC by Figure 11. The averages of the sulfate
545 concentrations obtained by NICAM-g6 tend to be smaller than those by NICAM-g6str
546 and the observations. The possible impacts of the prescribed oxidant on the sulfate
547 concentrations are investigated in section 3.2.2.

548 In Figure 13, NICAM-g6str and NICAM-g6 simulated SO₂ concentrations are
549 compared by the observations. In the previous studies, the comparison in SO₂
550 concentrations between the simulation and observation was very limited, with the
551 exception of their Figure 4 of Morino et al. (2010b), which showed large differences in
552 the SO₂ concentrations between WRF-CMAQ and the observations by more than a
553 factor of two. The R between NICAM-g6str and the observations are low, with the
554 exception of Komae (R=0.62), but are approximately within the range obtained by
555 WRF-CMAQ in Morino et al. (2010b). The differences in the mean SO₂ concentrations
556 between NICAM-g6str and the observations and between NICAM-g6 and the
557 observations are within approximately 20% at all sites, with the exception of
558 NICAM-g6str at Maebashi and NICAM-g6str at Tsukuba (Table 2). The temporal
559 variations in the simulated SO₂ concur with those in the observations. The observations
560 sometimes show high SO₂ concentrations at all sites, e.g., up to 20 ppbv at Komae, in
561 the afternoon on August 12 and 14. On August 12, NICAM-g6str normally reproduced

562 the peaks of the observed SO₂, but with the blunter and slightly shifted peaks. In the
563 NICAM-g6str simulation, the strong SO₂ plumes from industrial sources over the Tokyo
564 Bay arrived at the inner areas such as Kisai. On August 14, although the
565 NICAM-g6str-simulated winds were comparable to the observed ones (Figures 7 and 8),
566 NICAM-g6str did not reproduce the sharp peaks of the observed SO₂, especially at
567 Komae and Tsukuba. It may imply that special meteorological fields cause the observed
568 peaks on August 12, whereas unaccounted SO₂ emission from local sources or sporadic
569 volcanoes is stronger on August 14. The latter issue is improved by processing
570 time-highly-resolved emission inventories of SO₂, which can be estimated through a
571 top-down approach using a data assimilation (Schutgens et al., 2012; Xu et al., 2013).

572 To assess the performance of both NICAM-gs6tr and NICAM-g6 in simulating the
573 distributions of the air pollutants over Japan, we compared the August averages of the
574 simulated EC, sulfate and SO₂ concentrations with the available measurements (Figures
575 14 and 15). Although the EC observatories are limited, both the NICAM-g6str and
576 NICAM-g6 simulated EC concentrations are much underestimated compared to the
577 observations, with the relative bias (*Br*), defined as a ratio of the simulated value to the
578 observed one, to be 0.15 (NICAM-g6str) and 0.16 (NICAM-g6). In China, the
579 NICAM-g6str-simulated EC concentrations are comparable to the
580 NICAM-g6-simulated ones with the R values of 0.71 (NICAM-g6str) and 0.68
581 (NICAM-g6), whereas in Japan (no available measurements) the
582 NICAM-g6str-simulated EC concentrations are larger than NICAM-g6-simulated ones

583 at the Japanese urban areas such as Nagoya (136.97°E, 35.17°N) and Osaka (135.54°E,
584 34.68°N).

585 The NICAM-g6str-simulated sulfate concentrations are larger and more comparable to
586 the observations over China compared to NICAM-g6-simulated ones. In Japan, the hot
587 spots with greater concentrations of more than $5 \mu\text{g}/\text{m}^3$ are found only in the
588 NICAM-g6str results. The *Br* values are estimated to be 0.59 (NICAM-g6str) and 0.53
589 (NICAM-g6), whereas the *R* values are estimated to be 0.78 (NICAM-g6str) and 0.88
590 (NICAM-g6), respectively. The results indicate that the sulfate concentrations obtained
591 by both NICAM-g6str and NICAM-g6 tend to be underestimated by approximately
592 40-50% compared with the observed sulfate concentrations. The underestimation over
593 East Asia is mainly caused by the underestimation in China and possibly by the
594 uncertainty of the simulated precipitation around Japan. At Hedo located at Okinawa
595 islands, for example, the underestimation of both NICAM-g6str and NICAM-g6
596 simulated sulfate concentrations is caused by a possible underestimation of
597 transboundary sulfate from the continent, which is attributed to a large uncertainty of
598 the precipitation fields modulated by typhoon in the early August. However, the
599 correlations of sulfate between the simulations (both NICAM-g6str and NICAM-g6)
600 and observations are adequately acceptable. The simulated and observed SO_2
601 concentrations also correlate, with the *R* value of 0.63 (NICAM-g6str) and 0.48
602 (NICAM-g6). The *Br* values are calculated to be 0.48 (NICAM-g6str) and 0.67
603 (NICAM-g6). Figure 15 shows that the SO_2 , which is a primary product, is localized

604 near the source areas, whereas sulfate, which is as a secondary product, is distributed
605 from the source to the outflow areas. Although EC is also a primary product, the
606 horizontal distributions of NICAM-g6str-simulated EC are larger than those of
607 NICAM-g6str-simulated SO₂, possibly because EC is less scavenged through the dry
608 deposition and oxidation processes compared to SO₂.

609

610 **3.2.2 Uncertainty in the simulation**

611 Sensitivity tests were conducted to examine potential uncertainties derived from
612 prescribed datasets related to EC and sulfate for the NICAM-g6str simulations. For the
613 EC sensitivity tests, the emission quantities were set to half and twice of those used in
614 the standard run in this study. The results for the FAMIKA sites are shown in Figure
615 16(a) in which the bars show the simulated EC concentrations for both sensitivity tests.
616 For the majority of the sites, with the exception of Komae, the results obtained by the
617 sensitivity experiments of twice strength remain underestimated compared with the
618 measurements. The large underestimation of the EC mass concentrations at Maebashi
619 and Kisai was also shown by WRF-CMAQ of Shimadera et al. (2013) as well as the
620 previous studies of WRF-CMAQ in Morino et al. (2010a,b) and Chatani et al. (2014).
621 However, Fushimi et al. (2011) and Chatani et al. (2014) suggested that the difference
622 in the EC concentrations between WRF-CMAQ and the measurements is largely
623 attributed to an underestimation of the EC emission inventory, especially open biomass
624 burning from domestic sources. The local EC emission can be estimated by a

625 combination of the data assimilation and intensive measurements (Schutgens et al.,
626 2012; Wang et al., 2012; Yumimoto and Takemura, 2013).

627 Sensitivity experiments of the SO₂ emissions and the prescribed OH radical used in
628 sulfur chemistry were executed under half and twice the amounts used in the standard
629 experiment. The results for the FAMIKA sites are shown in Figure 16(b) in which the
630 bars show the simulated sulfate concentrations for both sensitivity tests under the
631 different experiments. Compared with the SO₂ emissions used in the standard
632 experiment, the doubled amount of SO₂ emissions can overcome the slight
633 underestimation of the simulated sulfate compared with the observations. Therefore, the
634 emission inventories of SO₂ should be improved for the better simulation of the sulfate.

635 In this sensitivity tests for oxidants, the SO₂ oxidation by OH radical strongly depends
636 on the OH concentrations as well as the cloud cover area, whereas the SO₂ oxidation by
637 ozone and hydrogen peroxide mainly depends on their concentrations, the cloud cover
638 area, and the cloud water content. The cloud distributions are modulated by some
639 feedbacks of the sulfate formation through the aerosol direct and indirect effects. As a
640 result, the sensitivity of the OH radical concentrations to the simulated sulfate
641 concentration is smaller than that we expected and that to the SO₂ emissions. We also
642 determined that the sensitivities of the other oxidants to the simulated sulfate
643 concentrations were small (not shown). These results and Figures 14 and 15 also
644 suggest that the use of the prescribed oxidants for sulfate formation is not crucial for
645 predicting monthly- and weekly-averaged sulfate mass concentrations at least by taking

646 into account for diurnal and seasonal variations of the prescribed oxidants. At the same
647 time, they also suggest that because the relationship between the oxidants and the
648 sulfate concentrations through the feedbacks is non-linear and complex, the use of the
649 prescribed oxidants for sulfate formation can affect the hourly variations of the sulfate
650 concentrations, and thus the sensitivity of the oxidants to the simulated sulfate should be
651 investigated.

652

653 **3.2.3 PM2.5**

654 Figure 17 shows the temporal variation in the surface PM2.5 mass concentration at the
655 18 Japanese sites including 10 sites in the Kanto area using different Y-axes for the
656 observed and simulated values. At most of the sites, both NICAM-g6str and NICAM-g6
657 usually captures the synoptic variation of the observed PM2.5, whereas only
658 NICAM-g6str reproduces the diurnal variation of the observed PM2.5. Table 3 shows
659 the PM2.5 concentrations in daily, daytime (from 9 am to 4 pm), and nighttime (from 9
660 pm to 4 am) averages and ratios of daytime to nighttime. The results show that the
661 simulated PM2.5 concentrations are underestimated compared with the observations by
662 more than a factor of two and by up to four at Maebashi. As for the diurnal variation,
663 the results show that the NICAM-g6str-simulated ratios (0.9-1.3) are larger than
664 NICAM-g6-simulated ones (0.8-0.9), whereas the NICAM-g6str-simulated ones are
665 smaller than the observed values (1.0-1.8). At Maebashi, where the ratio is higher than
666 that at other sites, the issue of the poor model performance of the meteorological fields

667 can be a major reason of the large underestimation, as mentioned in section 3.1. At all
668 sites, especially Maebashi and Kisai, the possible underestimation of SOA may be a
669 critical issue, as shown in the fact that the clear diurnal variation of PM_{2.5} during
670 August 4-9 and the high value of the ratios of daytime to nighttime and suggested by
671 previous studies (Matsui et al., 2009; Morino et al., 2010c). Morino et al. (2010c)
672 implied that over the Kanto area SOA from anthropogenic sources, which were
673 disregarded in this study, are large portion of total carbonaceous aerosols, even though
674 WRF-CMAQ does not correctly reproduce such carbonaceous aerosols. More
675 sophisticated SOA module, e.g., volatility basis-set approach proposed by Donahue et al.
676 (2006) based on the categorization of organic vapors with similar volatility, is required
677 for to produce SOA with higher accuracy. Originally, the underestimation of PM_{2.5} is
678 common among previous studies that employed regional aerosol-transport models
679 (Morino et al., 2010b, Chatani et al., 2011), primarily because the concentrations of the
680 observed PM_{2.5} include undefined chemical species with mean fractions ranging from
681 approximately 30–50% in the total PM_{2.5} in the summer of Japan (datasets from the
682 Tokyo Environment Agency and the Kawasaki Municipal Research Institute for
683 Environmental Protection). Another possible reason is that the PM_{2.5} mass
684 concentration includes water attached to aerosols, depending on the ambient RH
685 conditions. Therefore, these undefined chemical compounds in this study may account
686 for a large portion of the difference between the simulated and the observed values.
687 To evaluate the vertical profiles of the PM_{2.5} mass concentrations, we used the LIDAR

688 observation operated by the NIES-Japan network. Figure 18 shows the average results
689 for the simulated and observed extinction coefficient of the spherical particles at
690 Tsukuba and Chiba in August. At both sites, the vertical profiles and the magnitudes
691 below 3 km height of the simulated extinction by both NICAM-g6str and NICAM-g6
692 are comparable to the observed results, whereas the simulated extinction values tend to
693 be smaller than the observed extinction values near the surface. These results near the
694 surface are consistent with those obtained by the surface PM_{2.5} comparison shown in
695 Figure 17. In contrast, the extinction values observed by LIDAR include large
696 variabilities, primarily because they are retrieved from the surface to the cloud base,
697 which highly varies hour-by-hour and is basically difficult to detect with the high
698 accuracy, and secondly because they depend not only on the PM_{2.5} mass concentrations
699 but also on the ambient RH and the water amount attached to aerosols. At both sites, the
700 differences in the extinction between NICAM-g6str and NICAM-g6 are small below 1
701 km height, whereas those are relatively large above 1 km height. The differences are
702 attributed to the differences in the primary particles, mainly carbonaceous aerosols,
703 between NICAM-g6str and NICAM-g6 (not shown). It means that it is attributed to the
704 difference in the vertical transport between different spatial resolutions. Therefore,
705 impacts of the difference in the spatial resolution on the distributions of both aerosols
706 and their precursors should be addressed in the future work.

707

708 **4 Summary**

709 An aerosol-coupled global nonhydrostatic model, which is based on the aerosol module
710 of Spectral Radiation-Transport Model for Aerosol Species (SPRINTARS) and the
711 global cloud-resolving model of Nonhydrostatic Icosahedral Atmospheric Model
712 (NICAM), with a horizontal resolution of approximately 10 km or less in the target
713 region, is proposed in the present study. Circulations over both the global and target
714 domains are solved with a single model, whose mesh size varies with fine meshes
715 covering the target region, to calculate meso-scale circulations in the study region. The
716 stretched global model requires lower computational costs to simulate atmospheric
717 aerosols with fine horizontal resolutions compared with the global uniform
718 nonhydrostatic model, whereas it may require higher computational costs compared
719 with the general regional models, because the stretched-grid system calculates inside
720 and outside the target domain. As opposed to the general regional models, the
721 stretched-grid system does require neither nesting techniques nor boundary conditions.

722 In this study, we developed the new-type regional model with a horizontal resolution of
723 approximately 10 km to simulate aerosols over Japan, especially in the megacities of the
724 Kanto area, including Tokyo. To evaluate the model performances in the stretched-grid
725 system (hereafter referred to as the “NICAM-g6str”), we also simulated
726 NICAM-SPRINTARS with the globally uniformed grid simulation in glevel-6
727 resolution (the horizontal resolution is set to 110 km and we call it “NICAM-g6”). Both
728 NICAM-g6str and NICAM-g6 well reproduce general circulations obtained by
729 reanalysis of NCEP-FNL under the nudging technique over Asia including the target

730 region. Only NICAM-g6str usually reproduces both diurnal and synoptic variations of
731 the observed weather (temperature, wind, and precipitation) around Japan. Both
732 NICAM-g6str and NICAM-g6 generally reproduce monthly mean distributions of the
733 observed sulfate and SO₂ over East Asia, with the high correlations of more than 0.5,
734 but the underestimation of the simulated concentrations by 40% (NICAM-g6str) and
735 50% (NICAM-g6). The underestimation is mainly caused by the underestimation in
736 China and possibly by the uncertainty of the simulated precipitation around Japan. In
737 the Kanto area, the results obtained by NICAM-g6str are much closer to the
738 observations compared to those obtained by NICAM-g6. Only NICAM-g6str succeeds
739 in simulating the wind patterns and the diurnal transitions around the center of the
740 Kanto area, although it is inadequate to simulate the wind patterns and the diurnal
741 transitions at some sites located at the edge of the Kanto area and surrounded on three
742 sides by mountains, e.g., Maebashi, mainly due to the insufficient horizontal resolution.
743 NICAM-g6str also generally reproduces both diurnal and synoptic variations of the
744 observed and/or a regional aerosol-transport model (WRF-CMAQ) simulated EC,
745 sulfate, and SO₂ concentrations, especially with their high correlation ($R > 0.5$) at
746 Komae/Tokyo. The standard and sensitivity experiments suggest that (1) emission
747 inventories of EC and SO₂ should be improved for the better simulation and (2) the use
748 of the prescribed oxidants for the sulfate formation is not crucial for predicting weekly
749 and monthly averaged sulfate mass concentrations at least if the diurnal and seasonal
750 variations of the prescribed oxidants are considered. As for PM_{2.5} simulations, only
751 NICAM-g6str captures both synoptic and diurnal cycles of PM_{2.5}, with the exception

752 of the underestimation of the simulated PM_{2.5} by at least twice, probably due to the
753 underestimation of secondary organic aerosol (SOA) from anthropogenic sources and
754 the high uncertainties of the measurements.

755 Therefore, this new seamless aerosol-transport model, which covers global to regional
756 scales, can be applied to regional simulations. It suggests that even the simplified
757 aerosol module (e.g., prescribed oxidants for sulfur chemistry) is applicable for the
758 regional simulation if the module is coupled to a dynamic core with high horizontal
759 resolution. To more accurately simulate areas around Japan and develop the simplified
760 aerosol module, we need to address the following objectives: (1) to increase the
761 horizontal resolution (less than 10 km) to properly resolve wind fields, which can
762 greatly influence the delivery of air pollution from Tokyo to subcities such as
763 Maebashi; (2) to accurately reproduce the cloud and precipitation fields caused by
764 thermal lows, for example, by applying the finer horizontal resolution and/or more
765 sophisticated schemes of cloud microphysics such as the double-moment bulk scheme
766 proposed by Seiki and Nakajima (2014); (3) to use better emission inventories by
767 developing a data assimilation such as the Kalman smoother proposed by Schutgens et
768 al. (2012) with intensive measurements in many sites; (4) to simulate strong peaks of
769 PM_{2.5} in the daytime in the Kanto region by implementing more sophisticated module
770 of SOA formed from both anthropogenic and biogenic sources, such as the volatility
771 basis-set approach proposed by Donahue et al. (2006), in this model; and (5) to treat
772 nitrate aerosol through a thermodynamic equilibrium in the simulation of wintertime

773 and/or future scenarios where the relative contribution of nitrate will be larger than that
774 of sulfate under the changes in emission of NO_x and SO₂ (e.g., Ohara et al., 2007).
775 These issues are directly connected to the further development of NICAM-SPRINTARS
776 in both regional and global simulations. Near the future, we will present scenario
777 experiments at regional scales of 10 km grids and/or address the issue of regional air
778 quality and its health impacts in densely populated megacities.

779

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798

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1115 chemical signature, regional haze distribution and comparisons with global aerosols,
1116 *Atmos. Chem. Phys.*, 12, 779-799, doi: 10.5194/acp-12-779-2012, 2012.

1117 Table 1. Statistical values (averages of the observation and simulations, correlation
 1118 coefficient R and root-mean-square-error $RMSE$) for meteorological fields using the
 1119 simulations (NICAM-g6str and NICAM-g6) and observations at seven sites during the
 1120 same period, as shown in Figures 4 to 7.

		Yokohama	Chiba	Tsuchiura	Adachi	Maebashi	Machida	Kisai
		Temperature						
Average [°C] and difference [°C] (vs. observati on) in bracket	Observation	27.9	30.1	28.1	29.7	29.1	29.1	27.9
	NICAM-g6str	26.9 (-1.1)	28.3 (-1.8)	28.3 (0.2)	27.3 (-2.3)	25.5 (-3.6)	25.9 (-3.2)	25.8 (-2.2)
	NICAM-g6	25.5 (-2.4)	26.2 (-3.9)	25.7 (-2.4)	25.5 (-4.1)	23.9 (-5.2)	25.5 (-3.6)	23.9 (-4.0)
R	NICAM-g6str	0.74	0.85	0.84	0.81	0.79	0.74	0.80
	NICAM-g6	0.76	0.67	0.79	0.78	0.71	0.77	0.75
RMSE [°C]	NICAM-g6str	1.9	2.3	1.9	3.0	4.3	3.9	3.0
	NICAM-g6	2.8	4.4	3.1	4.6	5.8	4.0	4.6
		RH						
Average [%] and difference [%] (vs. observati on) in bracket	Observation	73.5	79.0	73.3	75.4	73.7	75.9	71.4
	NICAM-g6str	83.6 (10.0)	77.5 (-1.5)	76.4 (3.0)	77.9 (2.5)	82.7 (9.0)	82.5 (6.6)	81.6 (10.1)
	NICAM-g6	92.2 (18.6)	92.4 (13.4)	93.4 (20.0)	92.2 (16.8)	95.5 (21.9)	92.2 (16.3)	95.5 (24.1)
R	NICAM-g6str	0.64	0.68	0.69	0.72	0.72	0.72	0.81
	NICAM-g6	0.73	0.59	0.79	0.82	0.71	0.74	0.76
RMSE [%]	NICAM-g6str	12.7	8.9	11.0	10.1	14.6	12.9	13.3
	NICAM-g6	19.5	16.2	22.4	19.8	25.5	20.1	26.3
		Wind speed						
Average [m/s] and	Observation	2.9	2.6	1.6	2.6	1.2	2.7	1.9
	NICAM-g6str	4.2	3.8	3.1	3.4	3.1	3.0	2.7

difference		(1.3)	(1.1)	(1.4)	(0.9)	(1.9)	(0.3)	(0.8)
[m/s] (vs. observation) in bracket	NICAM-g6	3.7 (0.7)	5.0 (2.4)	1.0 (-0.7)	3.7 (1.1)	0.9 (-0.4)	3.7 (1.0)	0.9 (-1.0)
R	NICAM-g6str	0.72	0.41	0.65	0.51	0.19	0.59	0.16
	NICAM-g6	0.64	0.43	0.38	0.47	0.12	0.53	0.04
RMSE [m/s]	NICAM-g6str	1.9	2.0	1.8	1.7	2.3	1.3	1.7
	NICAM-g6	1.4	3.0	1.2	1.7	0.7	1.7	1.4

1121

1122 Table 2. Statistical values (averages of the observation and simulations, correlation
1123 coefficient R and root-mean-square-error $RMSE$) for EC, sulfate, and SO_2
1124 concentrations by the simulations (NICAM-g6str, NICAM-g6, and WRF-CMAQ) and
1125 the observations at four FAMIKA sites during the period from August 6 to 11. The
1126 WRF-CMAQ results are given by Shimadera et al. (2013).

		Maebashi	Kisai	Komae	Tsukuba
		EC			
Average [$\mu\text{g}/\text{m}^3$] and difference [%] (vs. observation) in bracket	Observation	2.85	2.75	1.23	2.20
	NICAM-g6str	0.39 (-86)	0.60 (-78)	1.10 (-10)	0.73 (-67)
	NICAM-g6	0.52 (-82)	0.52 (-81)	0.49 (-60)	0.58 (-74)
	WRF-CMAQ	0.87 (-69)	1.17 (-58)	0.92 (-25)	0.77 (-65)
R	NICAM-g6str	-0.02	0.41	0.55	0.59
	NICAM-g6	-0.49	-0.28	-0.05	0.16
	WRF-CMAQ	0.08	0.33	0.37	-0.23
RMSE [$\mu\text{g}/\text{m}^3$]	NICAM-g6str	2.62	2.33	0.72	1.85
	NICAM-g6	2.52	2.45	1.10	2.06
	WRF-CMAQ	2.18	1.83	0.88	1.98
		Sulfate			
Average [$\mu\text{g}/\text{m}^3$] and difference [%] (vs. observation) in bracket	Observation	4.79 (-6)	2.86 (44)	4.18 (-32)	4.85 (-12)
	NICAM-g6str	4.51 (-34)	4.14 (11)	2.84 (-46)	4.25 (-26)
	NICAM-g6	3.17 (-21%)	3.17 (42%)	2.25 (-21%)	3.58 (-22%)
	WRF-CMAQ	3.77	4.08	3.30	3.80

R	NICAM-g6str	0.01	0.50	0.51	0.73
	NICAM-g6	0.05	0.56	0.86	0.75
	WRF-CMAQ	0.41	0.02	0.87	0.78
RMSE [$\mu\text{g}/\text{m}^3$]	NICAM-g6str	3.61	2.81	2.71	2.49
	NICAM-g6	3.01	2.30	2.49	2.77
	WRF-CMAQ	2.30	3.37	1.62	2.56
SO ₂					
Average [ppbv] and difference [%] (vs. observation) in bracket	Observation	2.74	2.28	2.35	3.79
	NICAM-g6str	1.25 (-54)	1.90 (-17)	2.34 (-1)	2.34 (-38)
	NICAM-g6	2.42 (-12)	2.45 (7)	2.52 (7)	3.21 (-15)
R	NICAM-g6str	0.02	-0.04	0.62	0.21
	NICAM-g6	-0.64	-0.52	0.22	-0.04
RMSE [ppbv]	NICAM-g6str	1.82	0.93	0.97	2.08
	NICAM-g6	1.29	0.94	0.85	1.29

1127

1128 Table 3. PM_{2.5} concentrations in daily, daytime (from 9 am to 4 pm), and nighttime
1129 (from 9 pm to 4 am) averages and mean ratios of daytime to nighttime using the
1130 simulations (NICAM-g6str and NICAM-g6) and the observation at selected seven sites
1131 in August.

	Maebashi	Kawasaki	Toride	Hasuda	Sapporo	Nagoya	Fukuoka
Daily mean PM _{2.5} [$\mu\text{g}/\text{m}^3$] and standard deviation [$\mu\text{g}/\text{m}^3$]							
Observation	24.9±12.8	23.2±12.9	17.6±9.7	20.6±11.5	12.7±6.3	17.3±10.1	14.3±7.5
NICAM-g6str	6.4±3.9	10.0±7.3	9.0±6.3	8.4±5.0	4.9±3.5	7.5±5.7	3.4±2.6
NICAM-g6	6.7±3.0	6.7±3.3	6.7±3.4	6.7±3.0	4.7±4.1	5.4±3.0	3.5±2.3
Daytime (9am-4pm) mean PM _{2.5} [$\mu\text{g}/\text{m}^3$] and standard deviation [$\mu\text{g}/\text{m}^3$]							
Observation	28.6±14.1	19.4±12.1	15.8±9.0	21.0±10.0	15.0±5.2	11.3±5.4	9.7±5.7
NICAM-g6str	5.9±3.8	7.1±4.3	6.8±4.4	7.2±4.5	5.3±2.8	3.5±2.3	1.6±0.8
NICAM-g6	5.0±1.7	4.0±2.1	4.0±2.4	4.4±1.9	7.4±4.5	2.4±0.9	1.4±0.5
Nighttime (9pm-4am) mean PM _{2.5} [$\mu\text{g}/\text{m}^3$] and standard deviation [$\mu\text{g}/\text{m}^3$]							
Observation	24.4±11.9	24.5±11.8	16.9±9.6	18.5±10.3	10.7±6.6	19.1±8.2	15.4±6.7
NICAM-g6str	7.5±3.6	14.2±9.2	12.1±7.6	10.8±5.5	4.1±3.9	12.0±4.6	5.1±3.1

NICAM-g6	7.5±2.3	9.1±1.5	8.8±2.1	8.4±3.0	2.6±3.1	7.8±1.3	4.4±2.2
	Ratio of daytime-mean PM2.5 to nighttime-mean PM2.5						
Observation	1.8±0.8	1.7±0.5	1.3±0.4	1.2±0.4	1.0±0.4	1.3±0.4	1.1±0.3
NICAM-g6str	1.1±0.6	1.3±0.7	1.1±0.6	1.1±0.5	0.9±0.3	1.2±0.9	1.0±0.6
NICAM-g6	0.9±0.2	0.8±0.1	0.8±0.1	0.8±0.1	0.8±0.2	0.9±0.2	0.8±0.2

1132

1133 **Figure captions**

1134 Figure 1 Topographical maps of (a) East Asia and (b) Eastern Japan, including the
1135 observation sites for the model validation. The topography is based on GTOPO30 (the
1136 horizontal resolution is 30 arc seconds, that is approximately 1 km) courtesy of the U.S.
1137 Geological Survey.

1138

1139 Figure 2 (a) EC and (b) SO₂ emission inventories in 2007.

1140

1141 Figure 3 Horizontal distributions of temperature and winds in August averages at the
1142 surface and the model height of approximately 5 km over Asia region using reanalysis
1143 data from NCEP-FNL, simulation by NICAM-g6str, and simulation by NICAM-g6.

1144

1145 Figure 4 Temporal variations in the NICAM-g6str and NICAM-g6 simulated and
1146 observed air temperature for a height of 2 m at (a) Yokohama, (b) Chiba, (c) Tsuchiura,
1147 (d) Adachi, (e) Maebashi, (f) Machida and (g) Kisai in August 2007.

1148

1149 Figure 5 Same as Figure 4 but for relative humidity (RH).

1150

1151 Figure 6 Same as Figure 4 but for wind direction.

1152

1153 Figure 7 Same as Figure 4 but for wind speed.

1154

1155 Figure 8 Horizontal distributions of precipitation in August averages derived from (a)
1156 simulation by NICAM-g6str, (b) simulation by NICAM-g6, (c) reanalysis data from
1157 MSM by JMA and (d) reanalysis data from GSMaP.

1158

1159 Figure 9 Temporal variations in the NICAM-g6str and NICAM-g6 simulated and
1160 observed precipitation amounts at 21 Japanese sites in August 2007. The comparison
1161 includes 10 sites in the Kanto area; (a) Maebashi, (b) Konosu, (c) Huchu, (d) Tsukuba,
1162 (e) Tokyo, (f) Yokohama, (g) Abiko, (h) Saitama, (i) Chiba, and (j) Nerima, 3 sites in
1163 the northern Japan; (k) Niigata, (l) Sendai, and (m) Sapporo, 5 sites in the western
1164 Japan; (n) Nagoya, (o) Osaka, (p) Himeji, (q) Fukuoka, and (r) Hyuga, and 3 remote
1165 islands (s) Hachijo-jima, (t) Oshima, and (u) Naha.

1166

1167 Figure 10 Predictive values of daily precipitation using the NICAM-g6str and
1168 NICAM-g6 simulations and the AMeDAS measurements during August 2007 at the
1169 sites defined at Figure 9, in units of percentage.

1170

1171 Figure 11 Temporal variations in the simulated (NICAM-g6str, NICAM-g6, and
1172 WRF-CMAQ) and observed EC mass concentrations near the surface at (a) Maebashi,
1173 (b) Kisai, (c) Komae and (d) Tsukuba in August 2007. The WRF-CMAQ results are
1174 given by Shimadera et al. (2013). The left axis in red represents the simulated values,
1175 and the right axis in black represents the observed values, in units of $\mu\text{g}/\text{m}^3$.

1176

1177 Figure 12 Same as Figure 11 but for sulfate.

1178

1179 Figure 13 Same as Figure 12 but for SO₂ without the WRF-CMAQ results, in units of

1180 ppbv.

1181

1182 Figure 14 Scatterplot of August mean concentrations for EC, sulfate and SO₂ between

1183 the simulations by NICAM-g6str and NICAM-g6 and the observations at the sites

1184 shown in the left panels. The statistics parameters, relative bias (Br) and correlation

1185 coefficient (R), calculated by the simulated and observed concentrations at all the sites,

1186 are also shown in each panel.

1187

1188 Figure 15 Horizontal distributions of concentrations for EC, sulfate and SO₂ near the

1189 surface using NICAM-g6str and NICAM-g6 in August averages. The circles in color

1190 shows the observation results at the sites.

1191

1192 Figure 16 (a) EC and (b) sulfate mass concentrations at the FAMIKA four sites using

1193 NICAM-g6str under the sensitivity experiments, WRF-CMAQ results shown by

1194 Shimadera et al. (2013) and the FAMIKA observations in averages of August 6-11. The

1195 bar represents the range of the sensitivity.

1196

1197 Figure 17 Temporal variations in the NICAM-g6str and NICAM-g6 simulated and

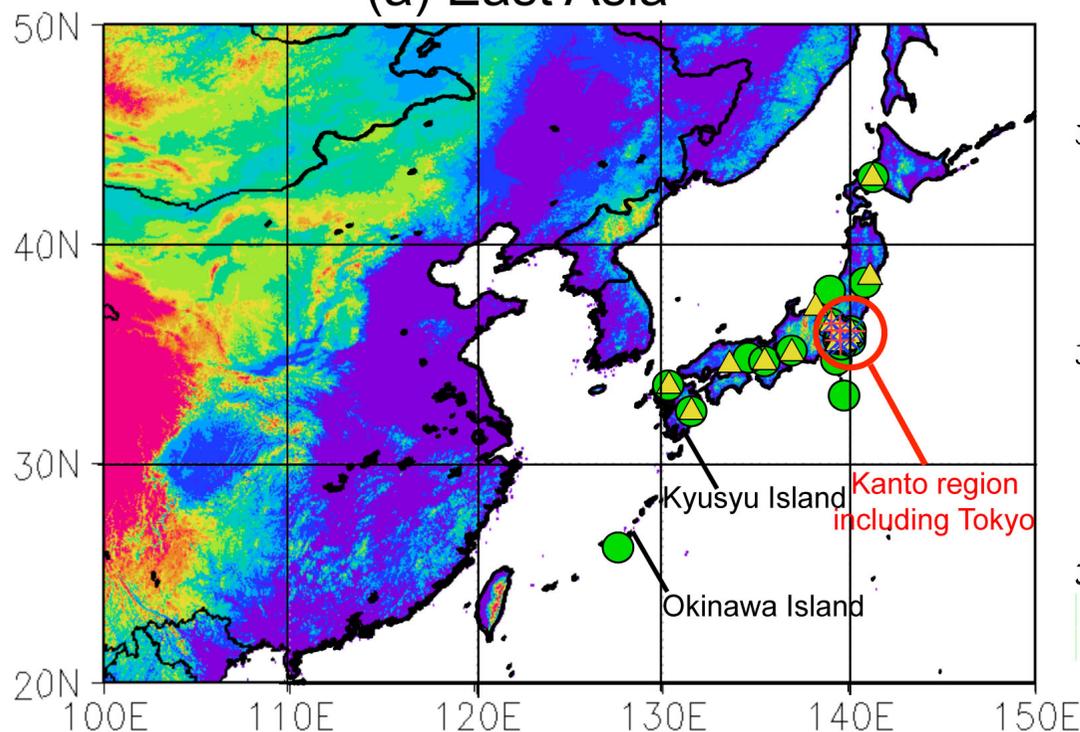
1198 observed PM_{2.5} near the surface at 18 Japanese sites in August 2007. The left axis in

1199 red represents the simulated values, and the right axis in black represents the observed
1200 values, in unit of μgm^{-3} .

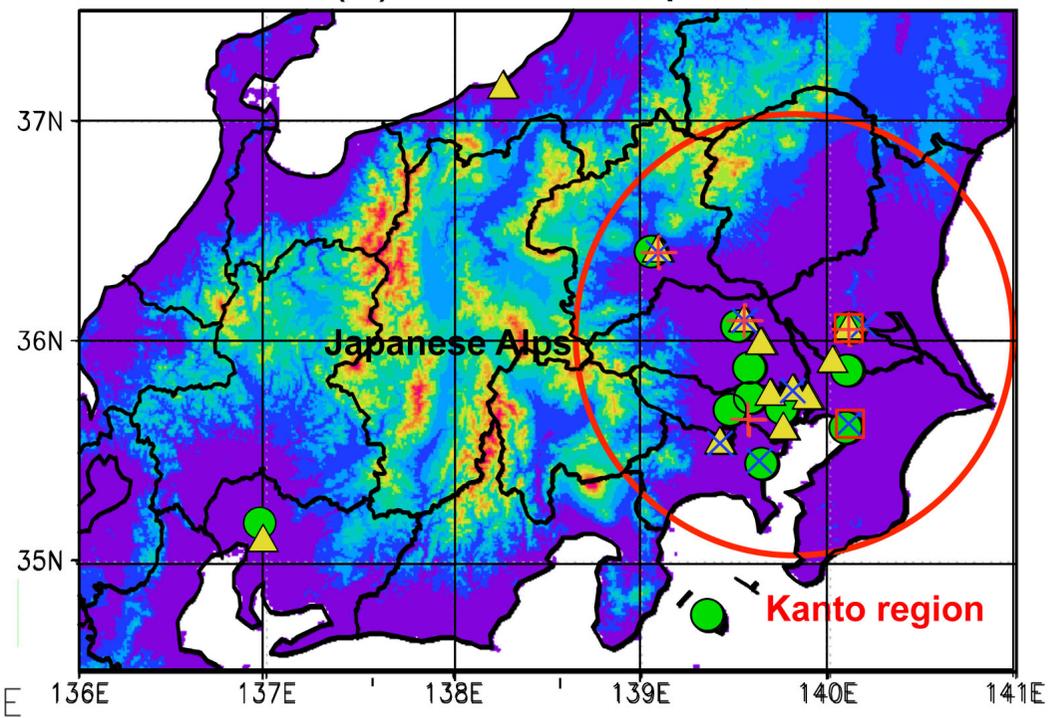
1201

1202 Figure 18 Extinction coefficients in August averages for the spherical particles
1203 simulated by NICAM-g6str and NICAM-g6 and the spherical particles observed by the
1204 NIES-LIDAR network at (a) Tsukuba and (b) Chiba, in units of $1/(\text{Mm})$. The bars
1205 represent the 25th and 75th percentiles of the LIDAR observations.

(a) East Asia

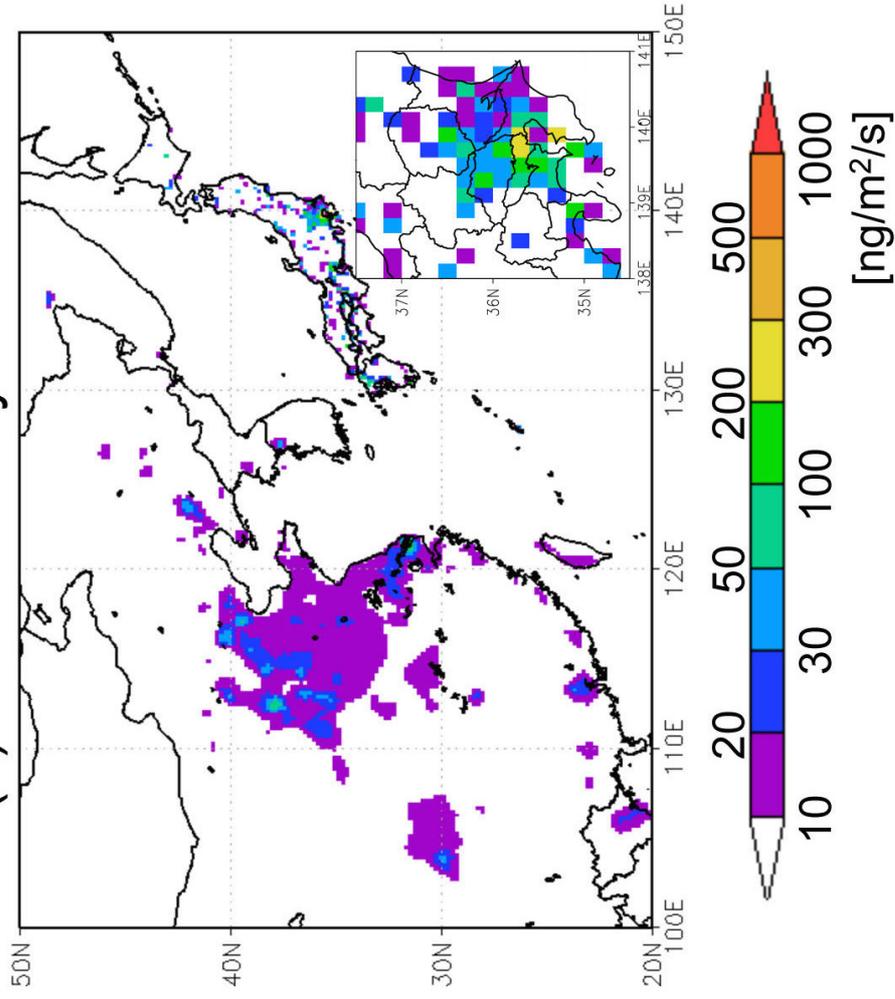


(b) Eastern Japan

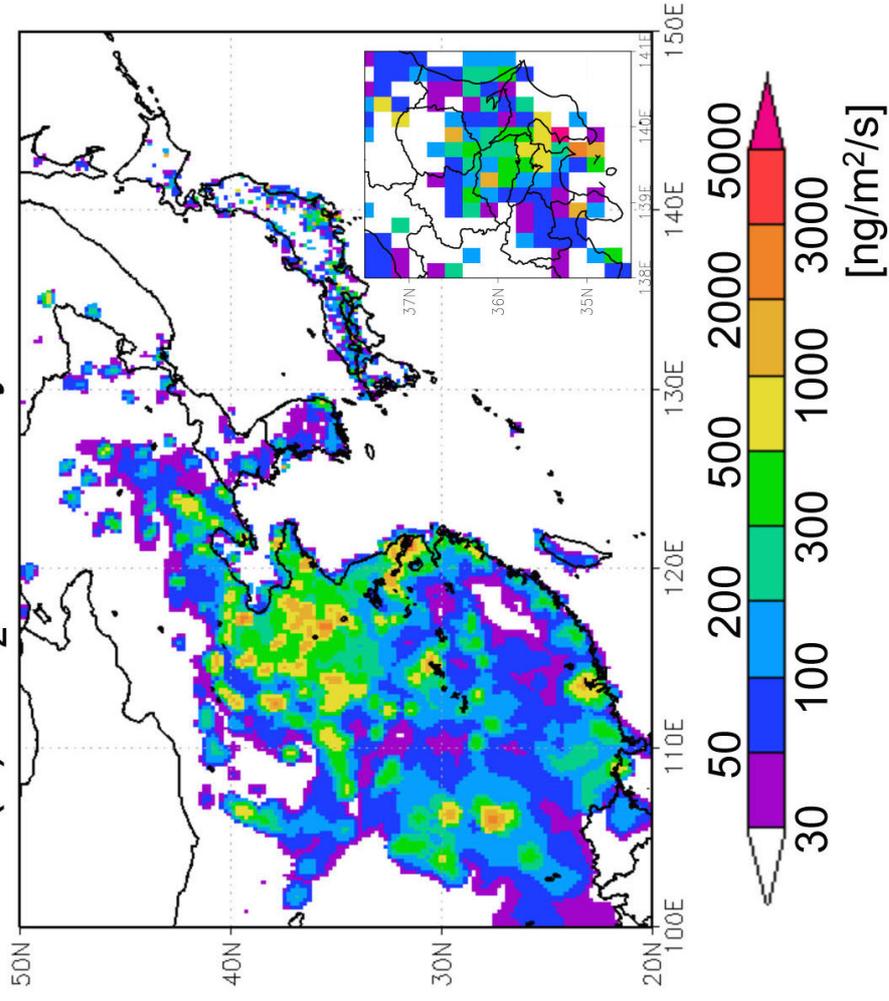


- + : FAMIKA sites (Aerosol chemical species) including 4 sites; Maebashi (139.10°E, 36.40°N), Kisai (139.56°E, 36.09°N), Komae (139.58°E, 35.64°N), Tsukuba (140.12°E, 36.05°N)
- × : 7 Sites (Meteorology measurements)
- : 21 Sites (Precipitation measurements by AMeDAS)
- ▲ : 18 Sites (PM_{2.5} measurements)
- : 2 Sites (LIDAR measurements)

(a) EC inventory in 2007

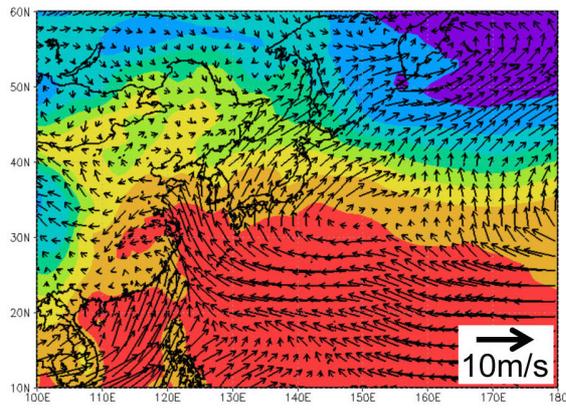


(b) SO₂ inventory in 2007

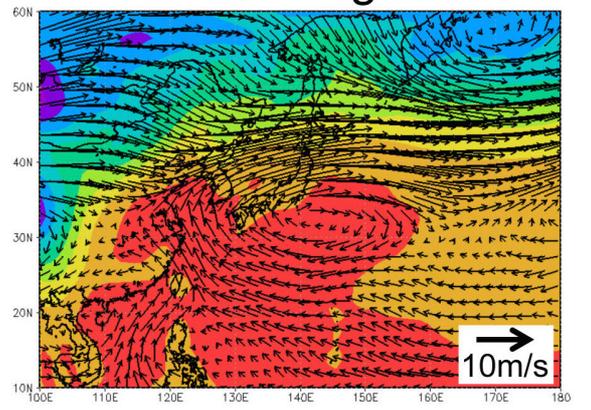


NCEP-FNL

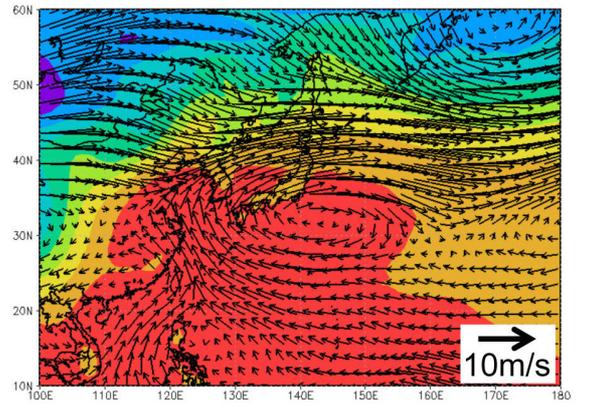
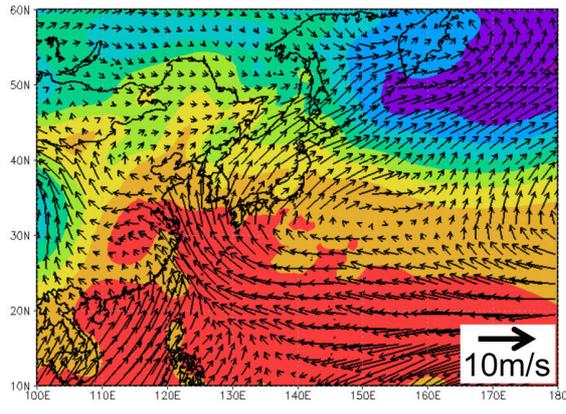
Surface



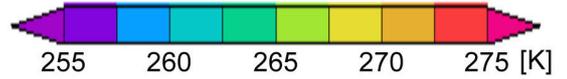
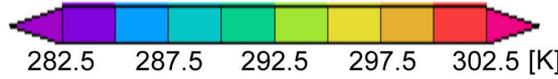
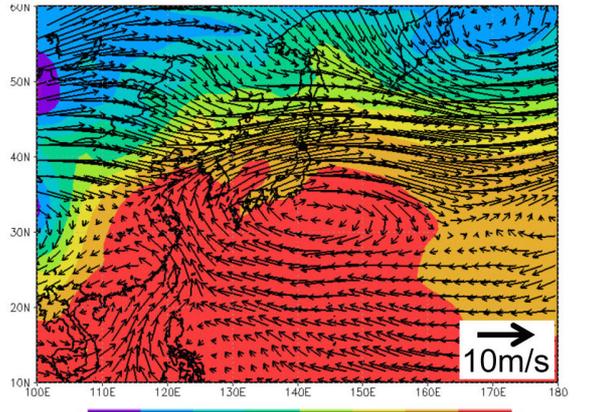
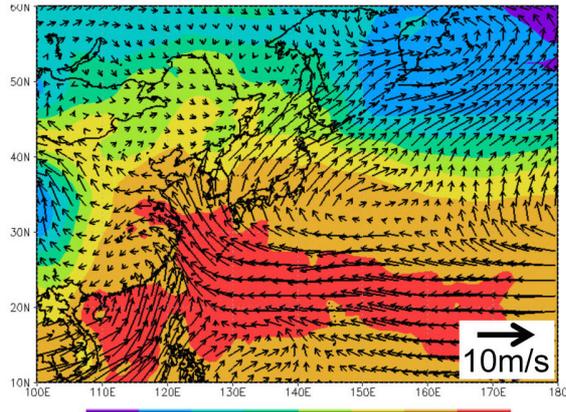
5km height



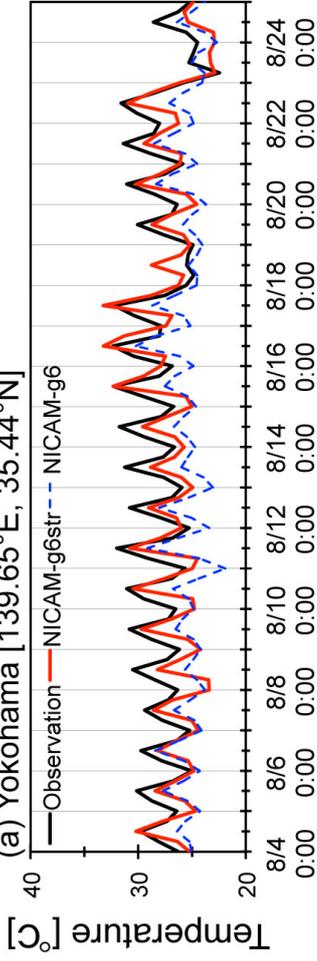
NICAM-g6str



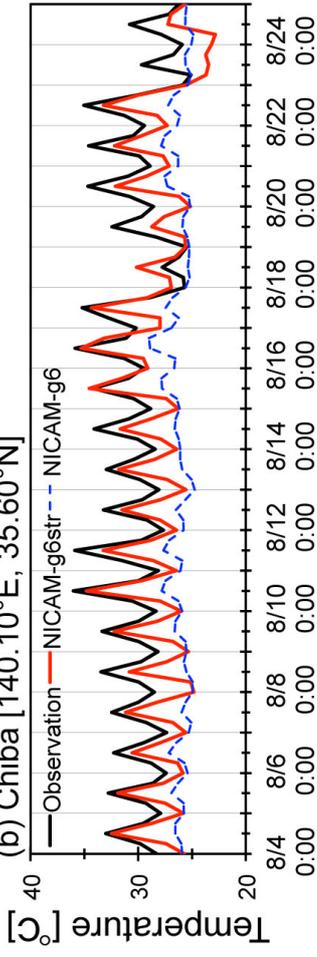
NICAM-g6



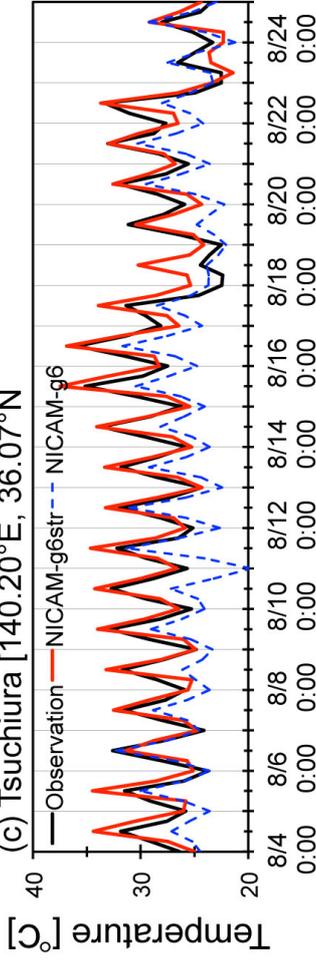
(a) Yokohama [139.65°E, 35.44°N]



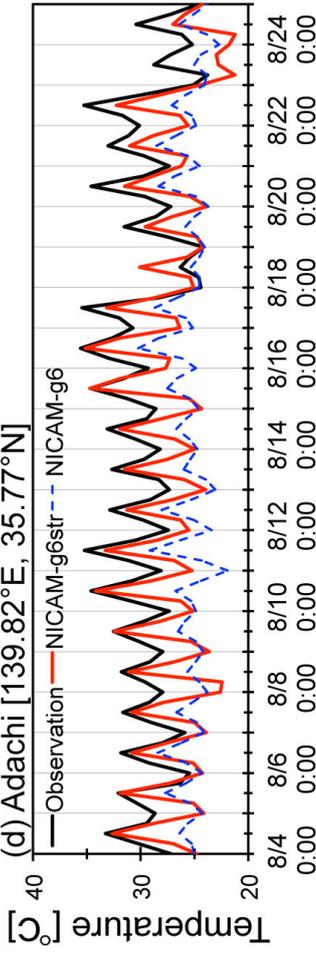
(b) Chiba [140.10°E, 35.60°N]



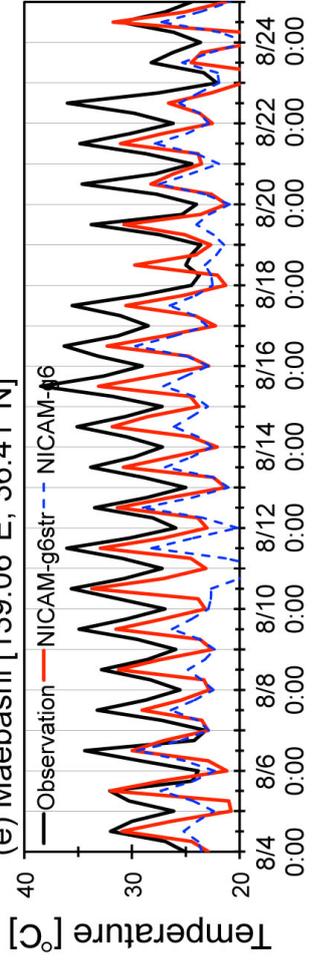
(c) Tsuchiura [140.20°E, 36.07°N]



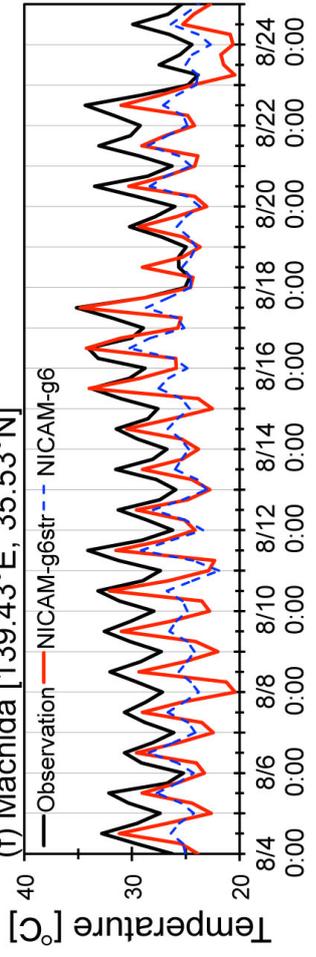
(d) Adachi [139.82°E, 35.77°N]



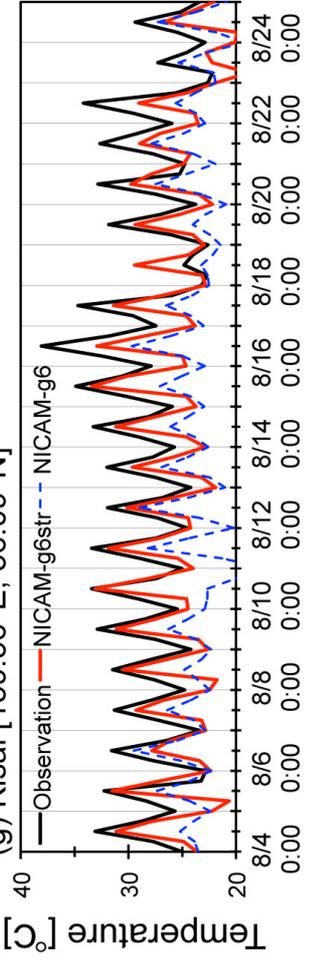
(e) Maebashi [139.06°E, 36.41°N]



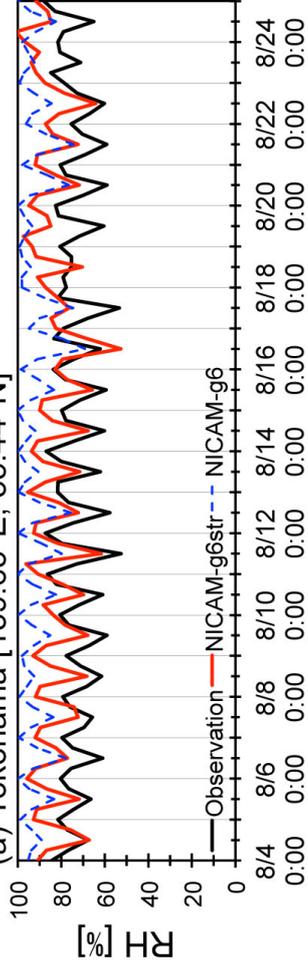
(f) Machida [139.43°E, 35.53°N]



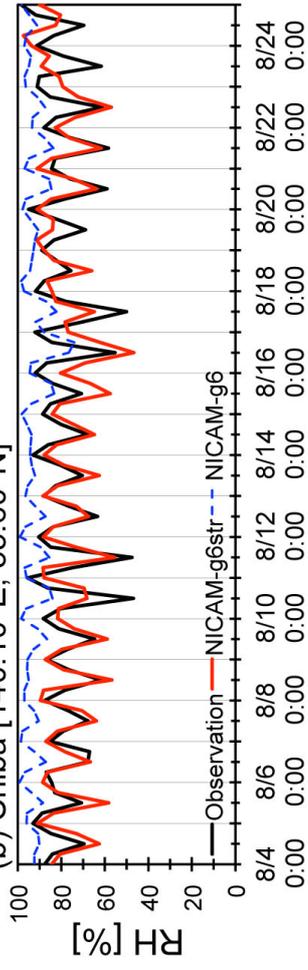
(g) Kisai [139.56°E, 36.09°N]



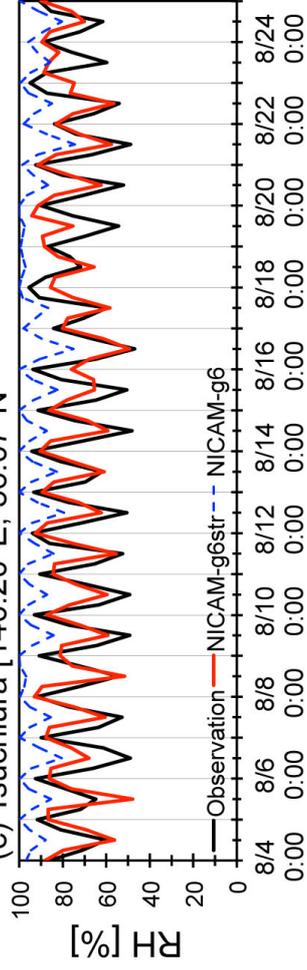
(a) Yokohama [139.65°E, 35.44°N]



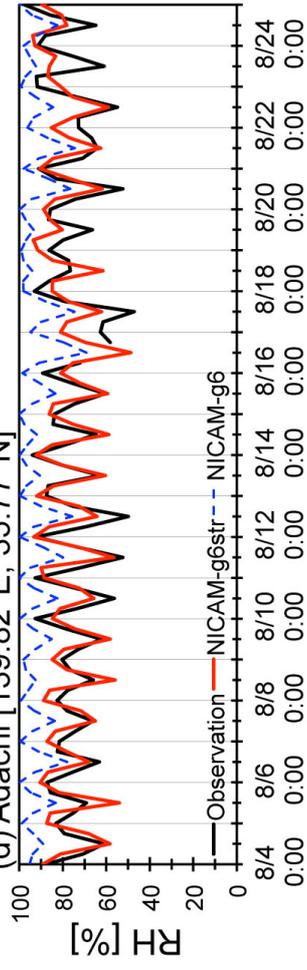
(b) Chiba [140.10°E, 35.60°N]



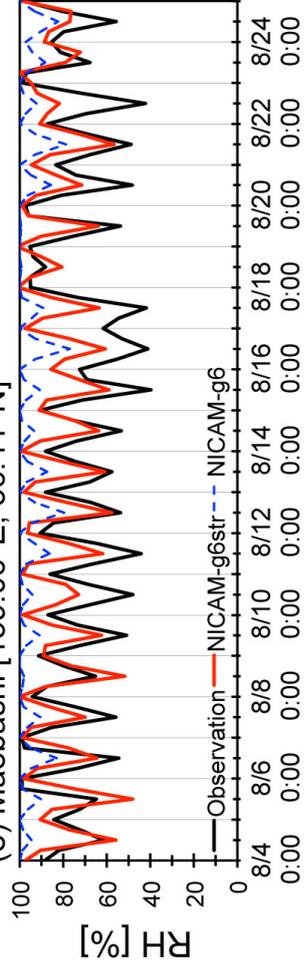
(c) Tsuchiura [140.20°E, 36.07°N]



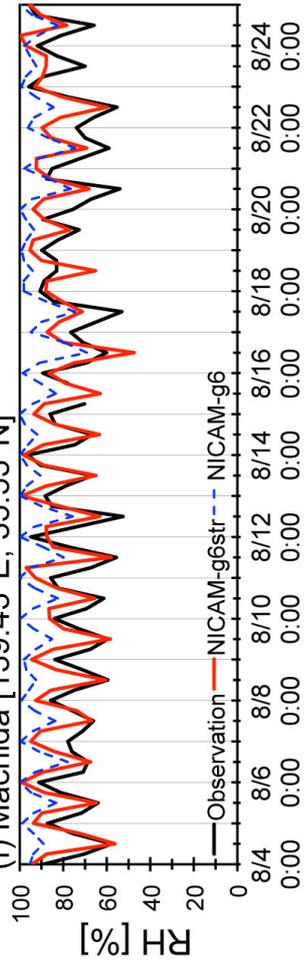
(d) Adachi [139.82°E, 35.77°N]



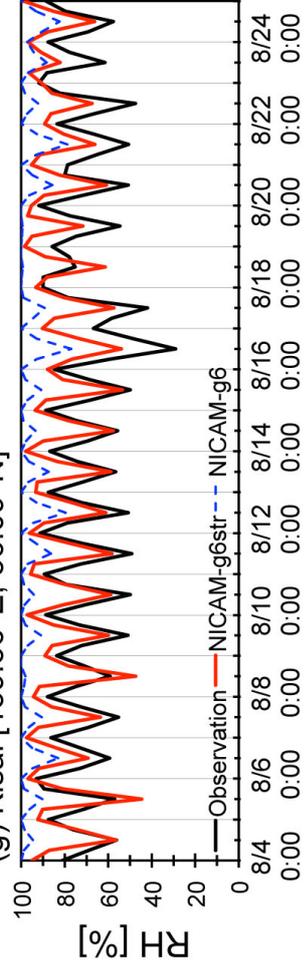
(e) Maebashi [139.06°E, 36.41°N]



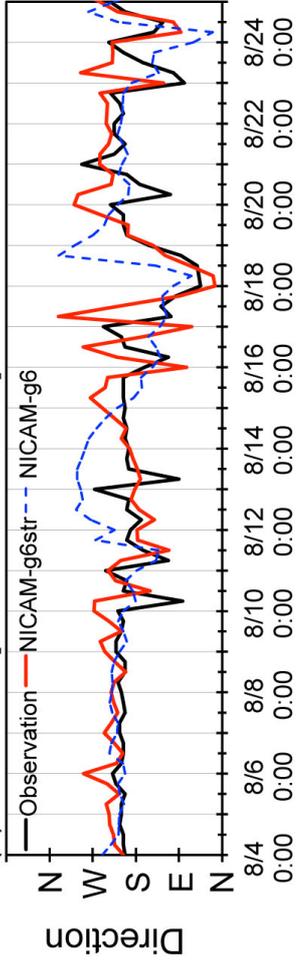
(f) Machida [139.43°E, 35.53°N]



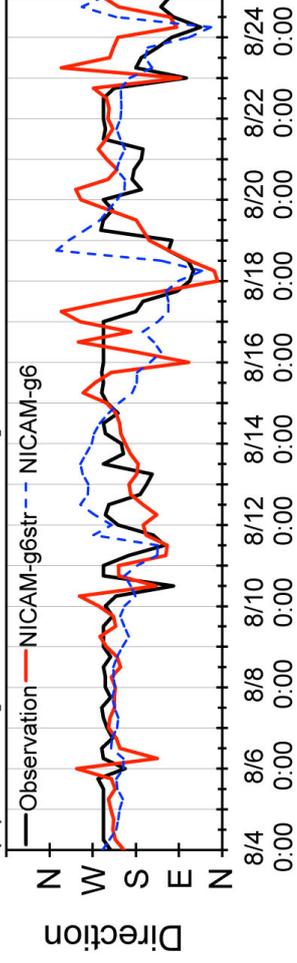
(g) Kisai [139.56°E, 36.09°N]



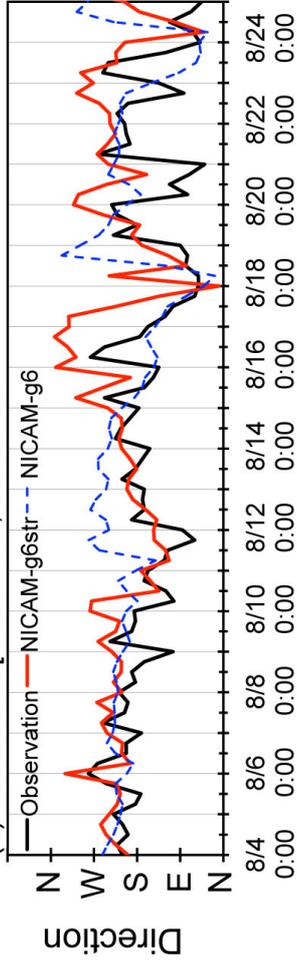
(a) Yokohama [139.65°E, 35.44°N]



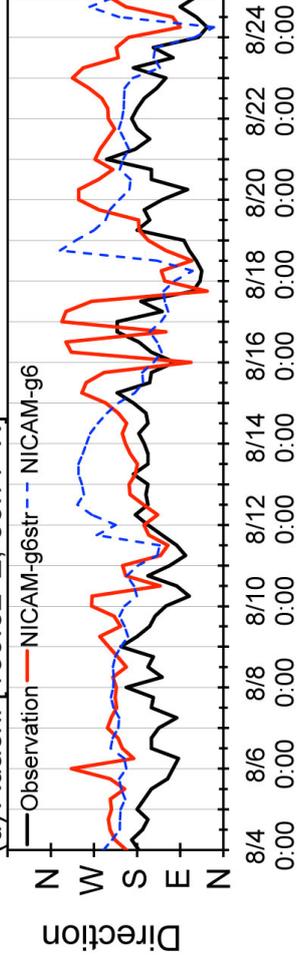
(b) Chiba [140.10°E, 35.60°N]



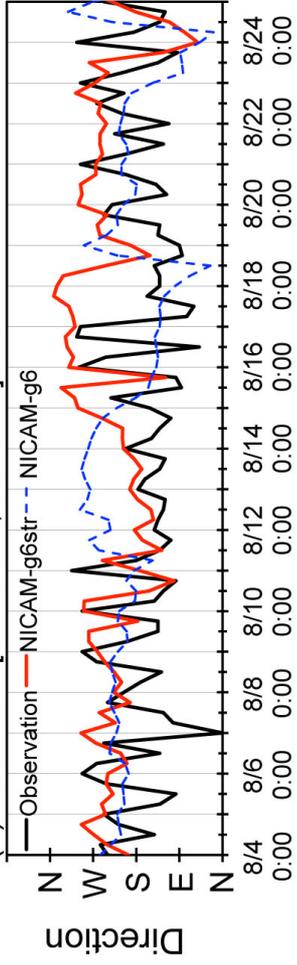
(c) Tsuchiura [140.20°E, 36.07°N]



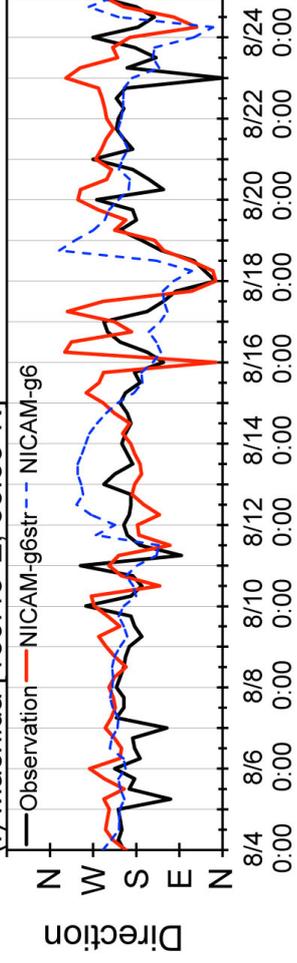
(d) Adachi [139.82°E, 35.77°N]



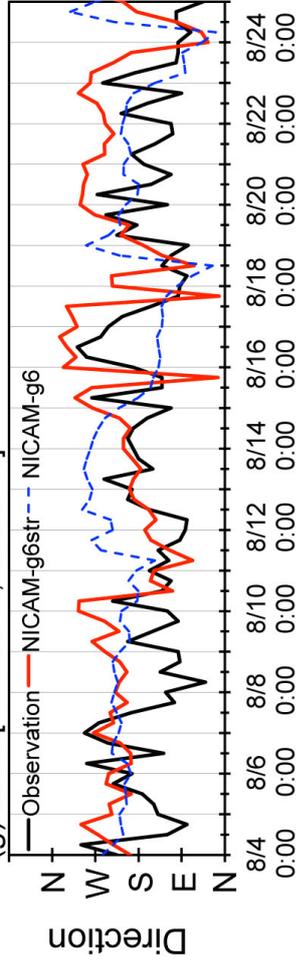
(e) Maebashi [139.06°E, 36.41°N]



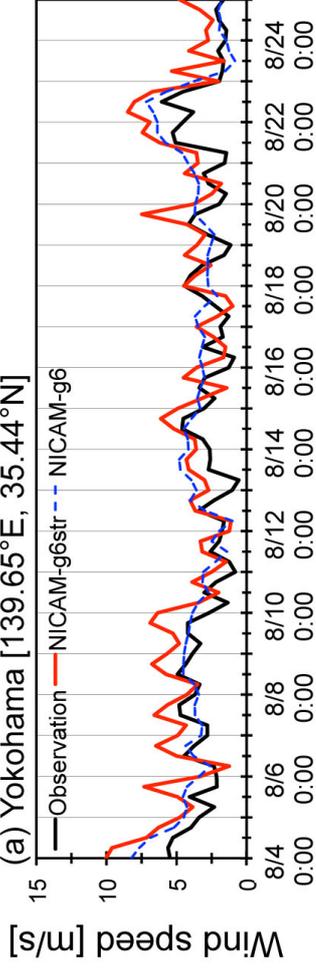
(f) Machida [139.43°E, 35.53°N]



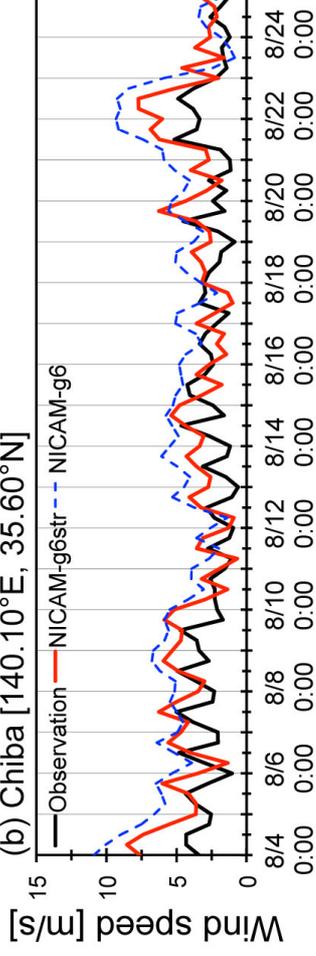
(g) Kisai [139.56°E, 36.09°N]



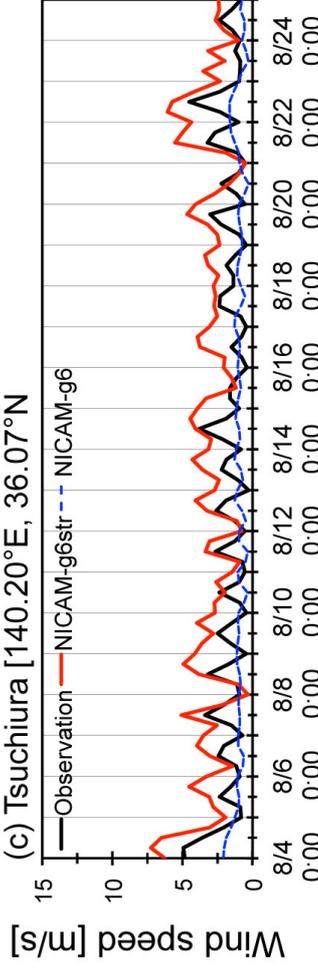
(a) Yokohama [139.65°E, 35.44°N]



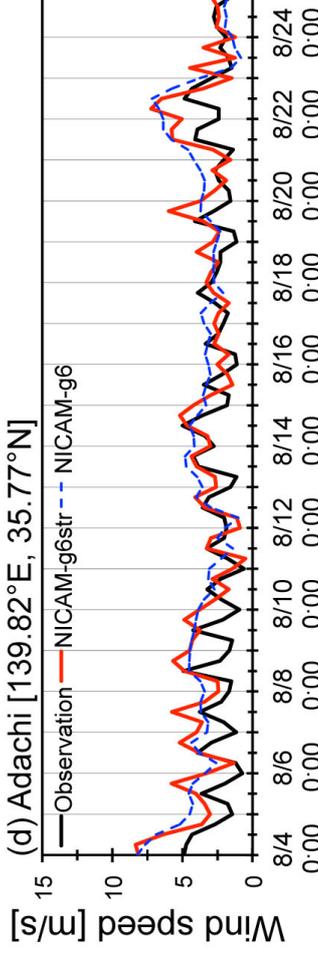
(b) Chiba [140.10°E, 35.60°N]



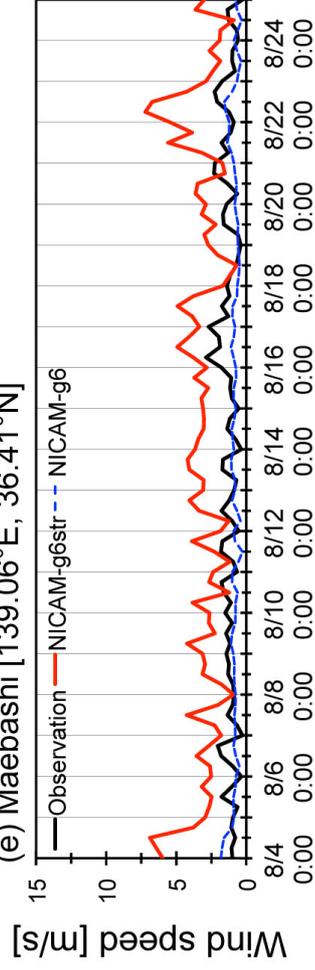
(c) Tsuchiura [140.20°E, 36.07°N]



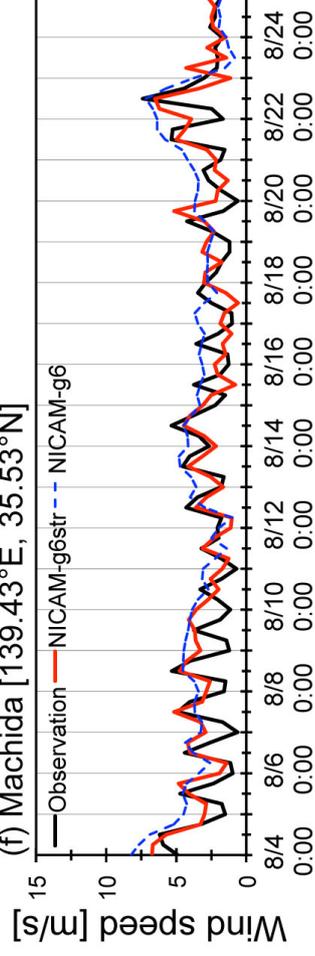
(d) Adachi [139.82°E, 35.77°N]



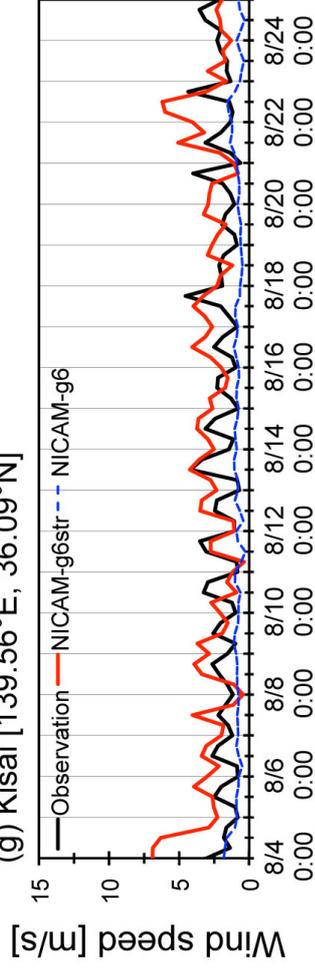
(e) Maebashi [139.06°E, 36.41°N]



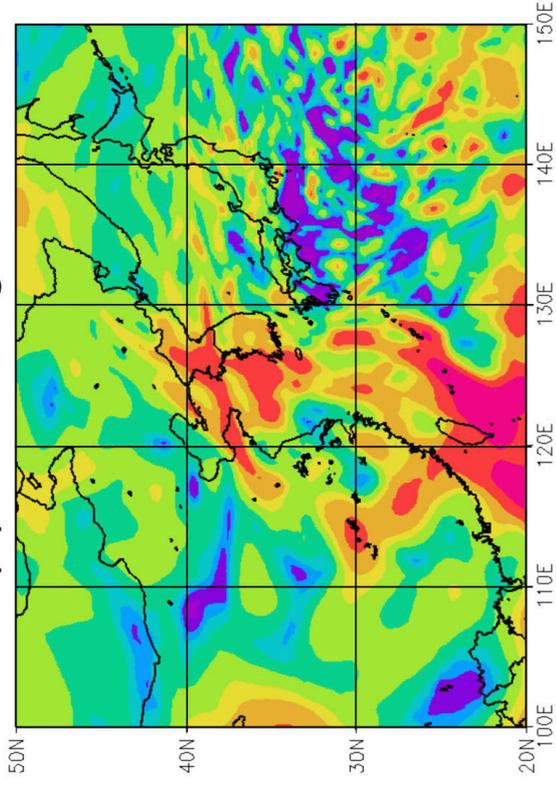
(f) Machida [139.43°E, 35.53°N]



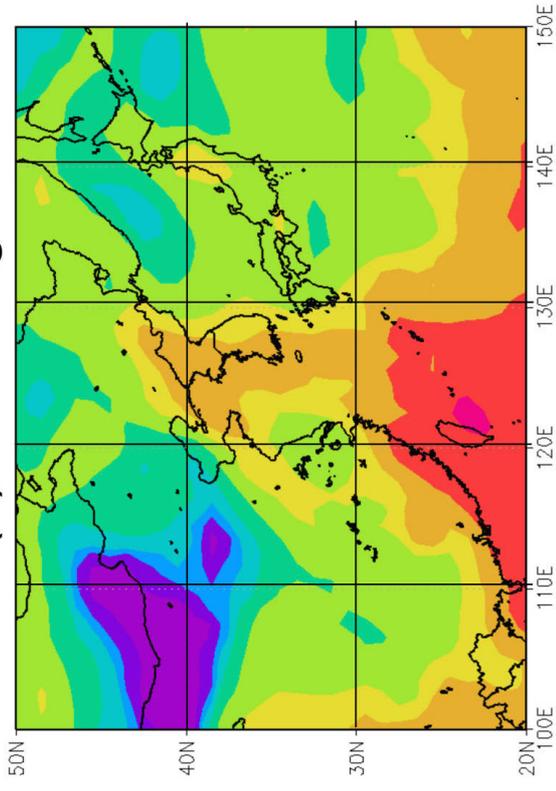
(g) Kisai [139.56°E, 36.09°N]



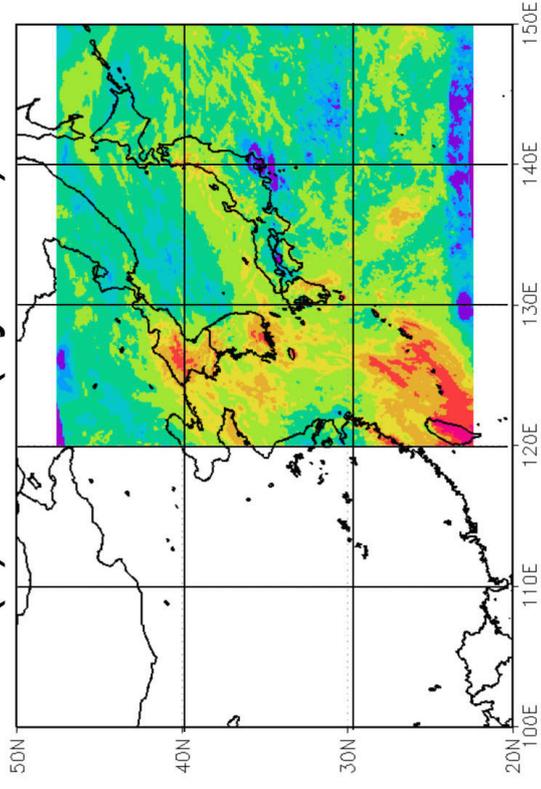
(a) NICAM-g6str



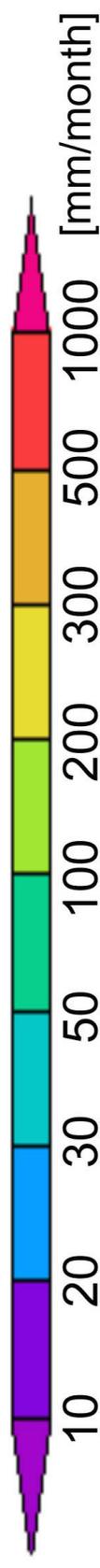
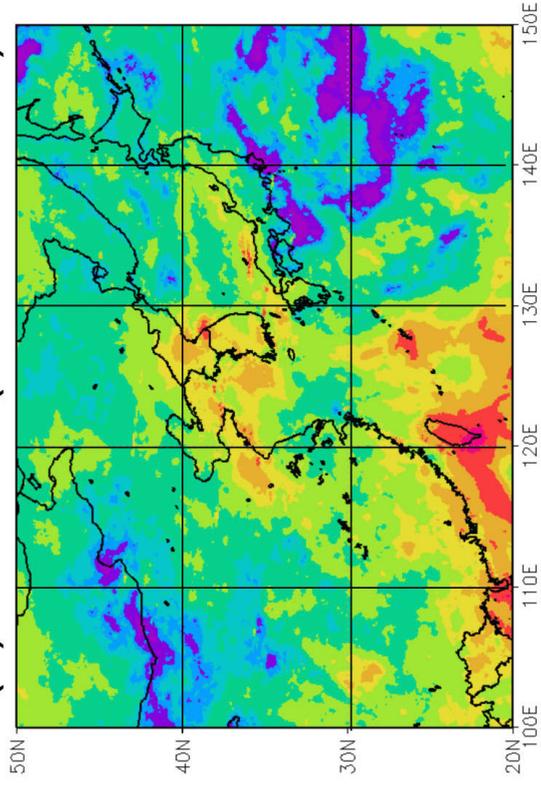
(b) NICAM-g6

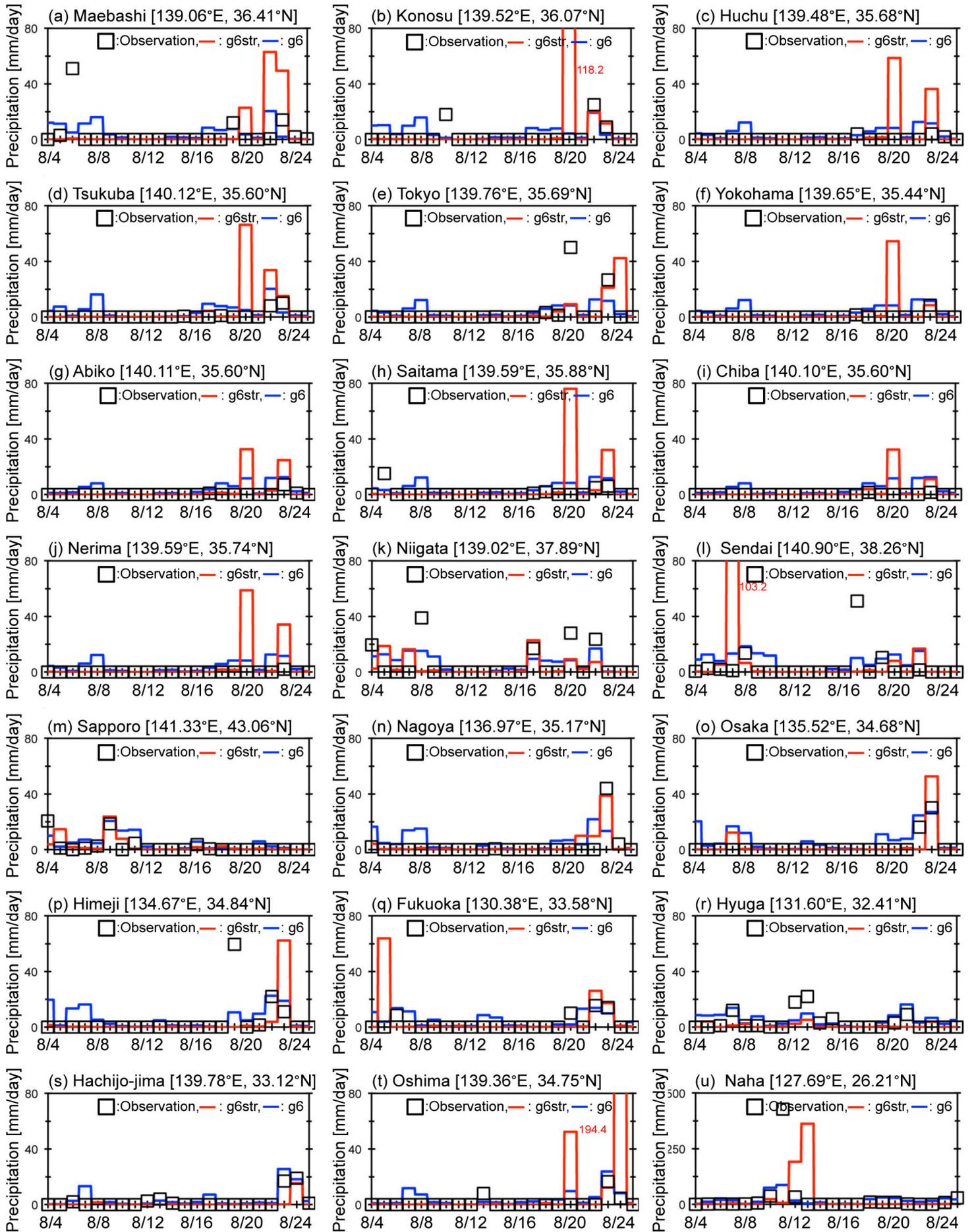


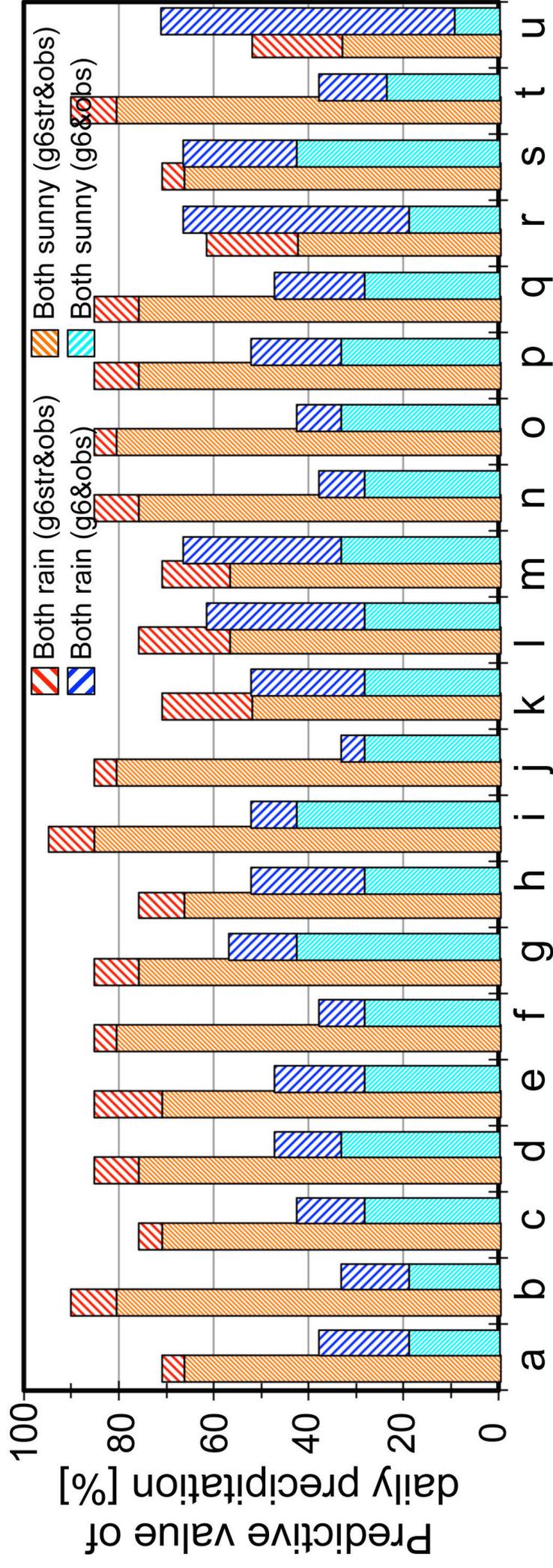
(c) MSM (by JMA)



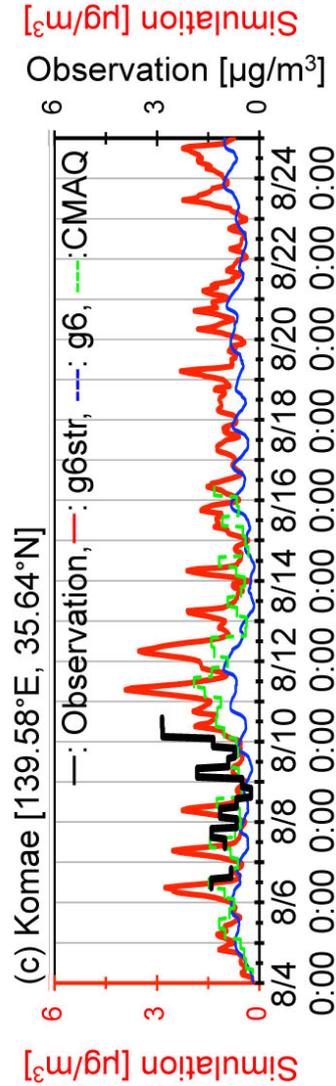
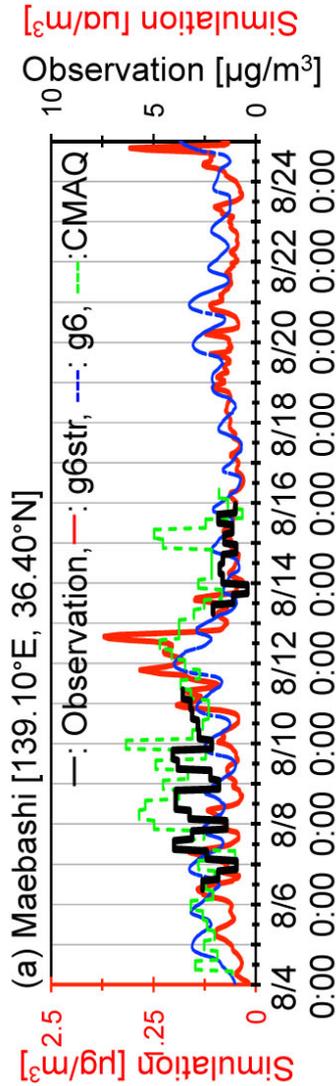
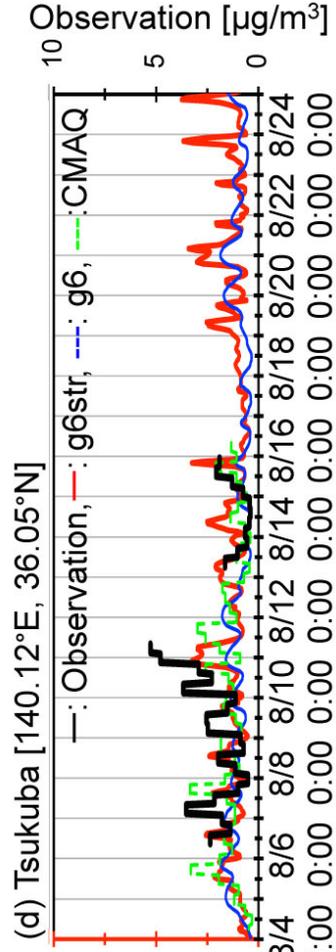
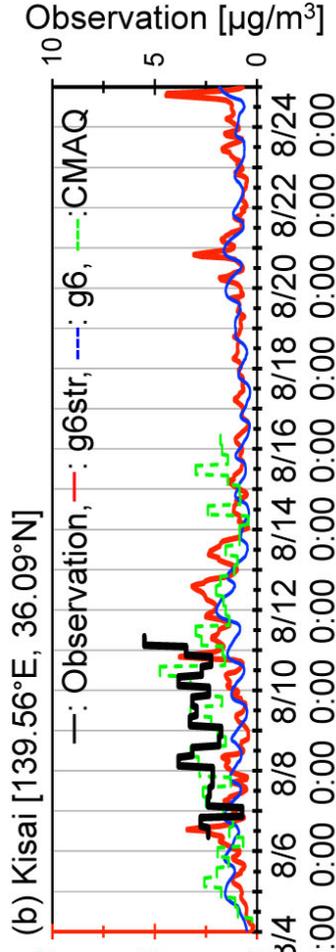
(d) GSMaP (multi-satellite)



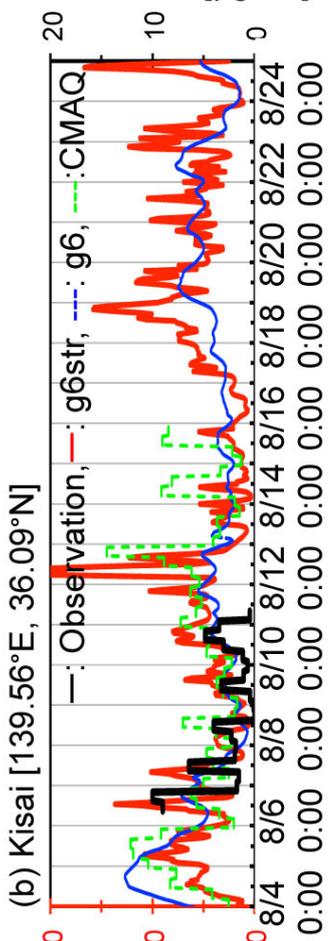




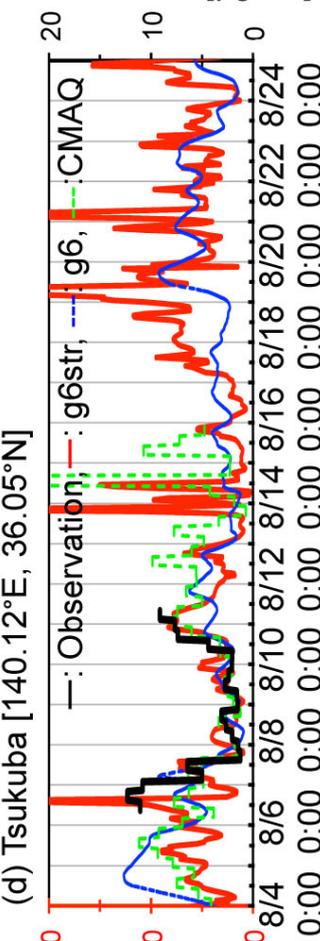
Sites (corresponding to the sites shown in Figure 9)



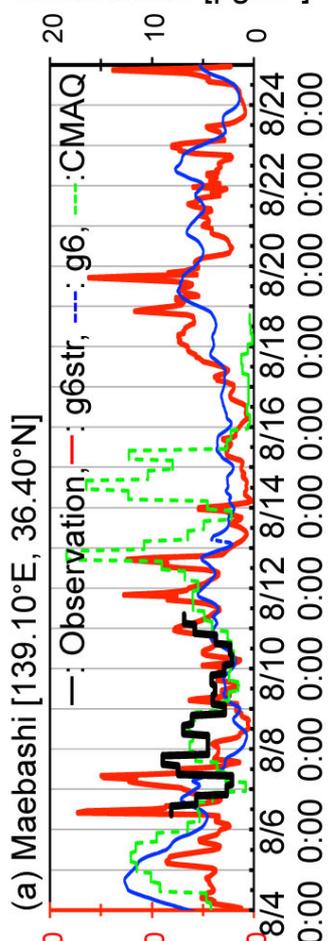
Observation [$\mu\text{g}/\text{m}^3$]



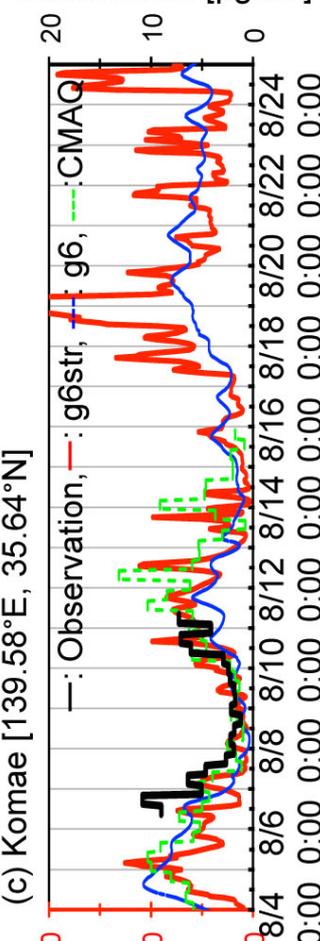
Observation [$\mu\text{g}/\text{m}^3$]



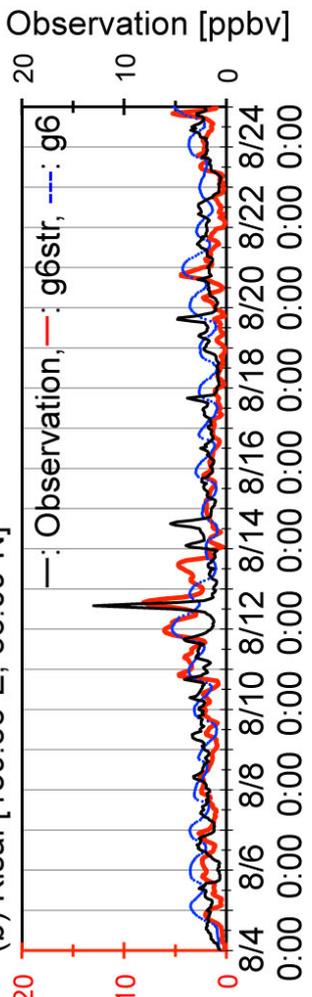
Observation [$\mu\text{g}/\text{m}^3$]



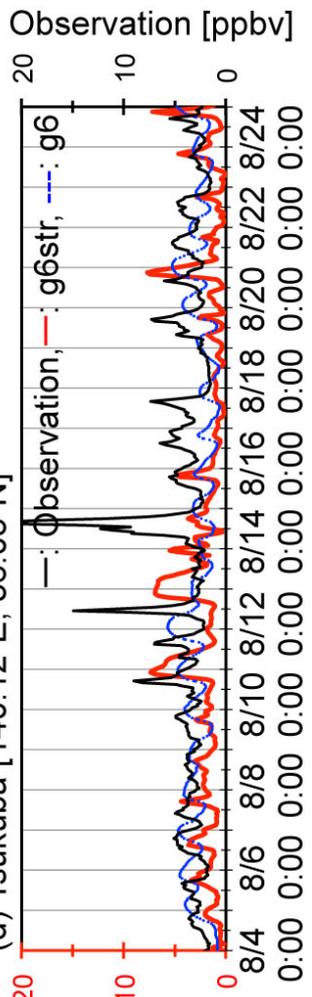
Observation [$\mu\text{g}/\text{m}^3$]



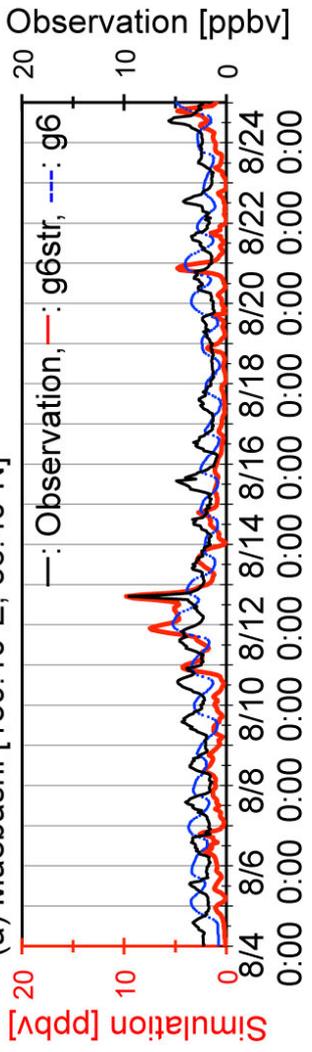
(b) Kisai [139.56°E, 36.09°N]



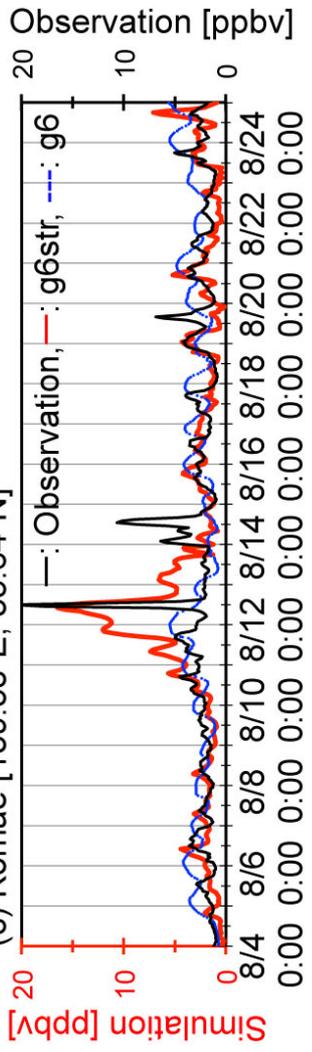
(d) Tsukuba [140.12°E, 36.05°N]

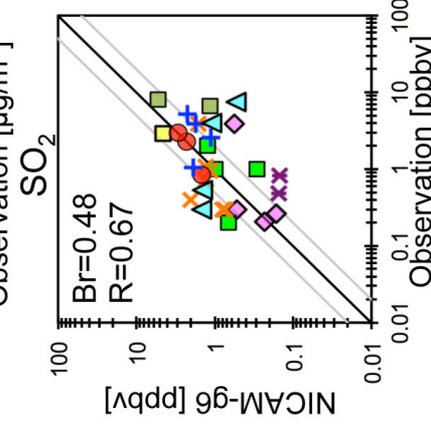
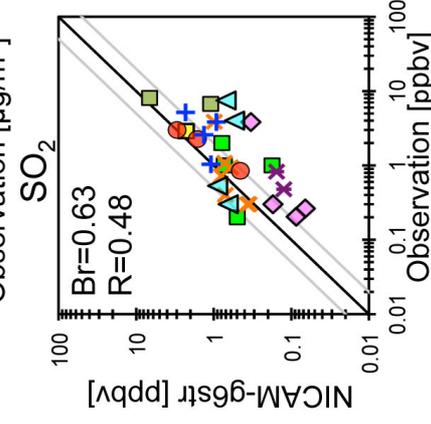
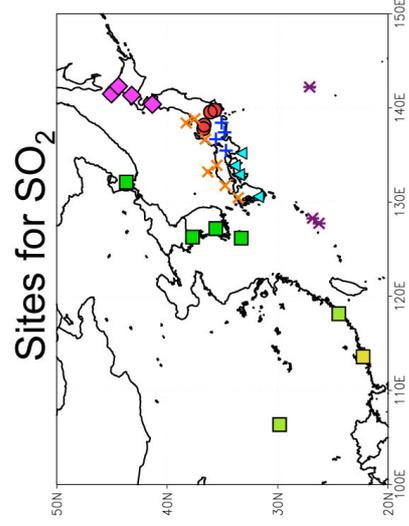
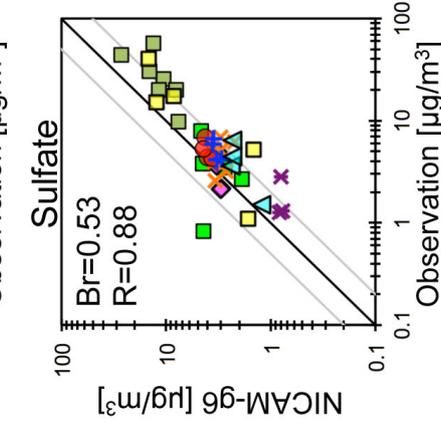
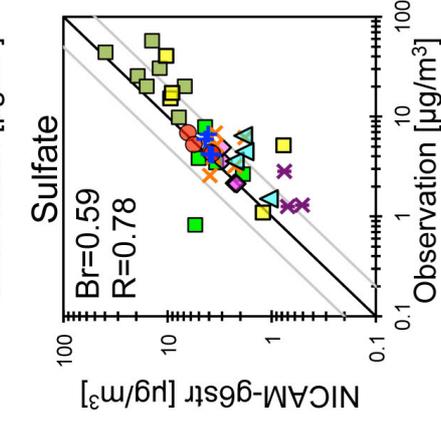
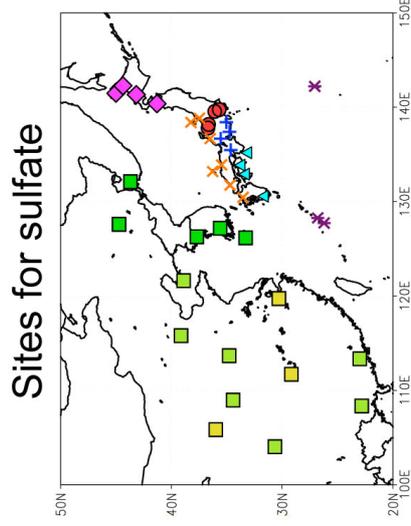
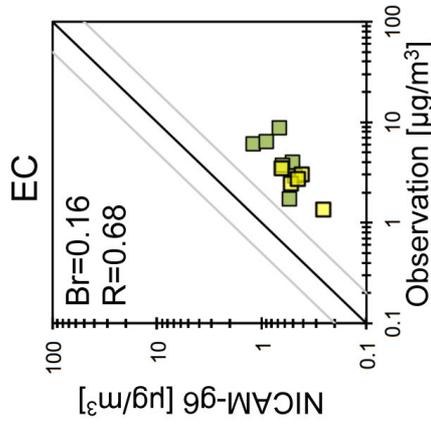
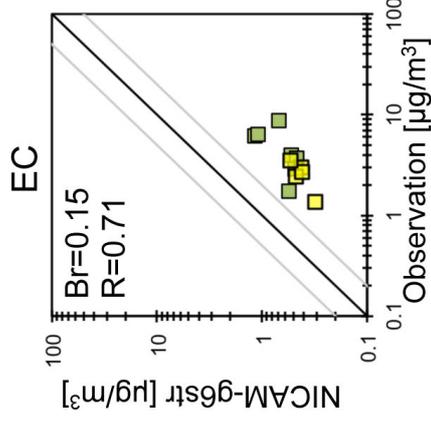
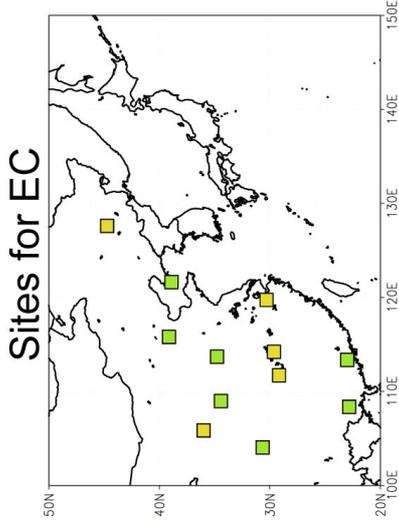


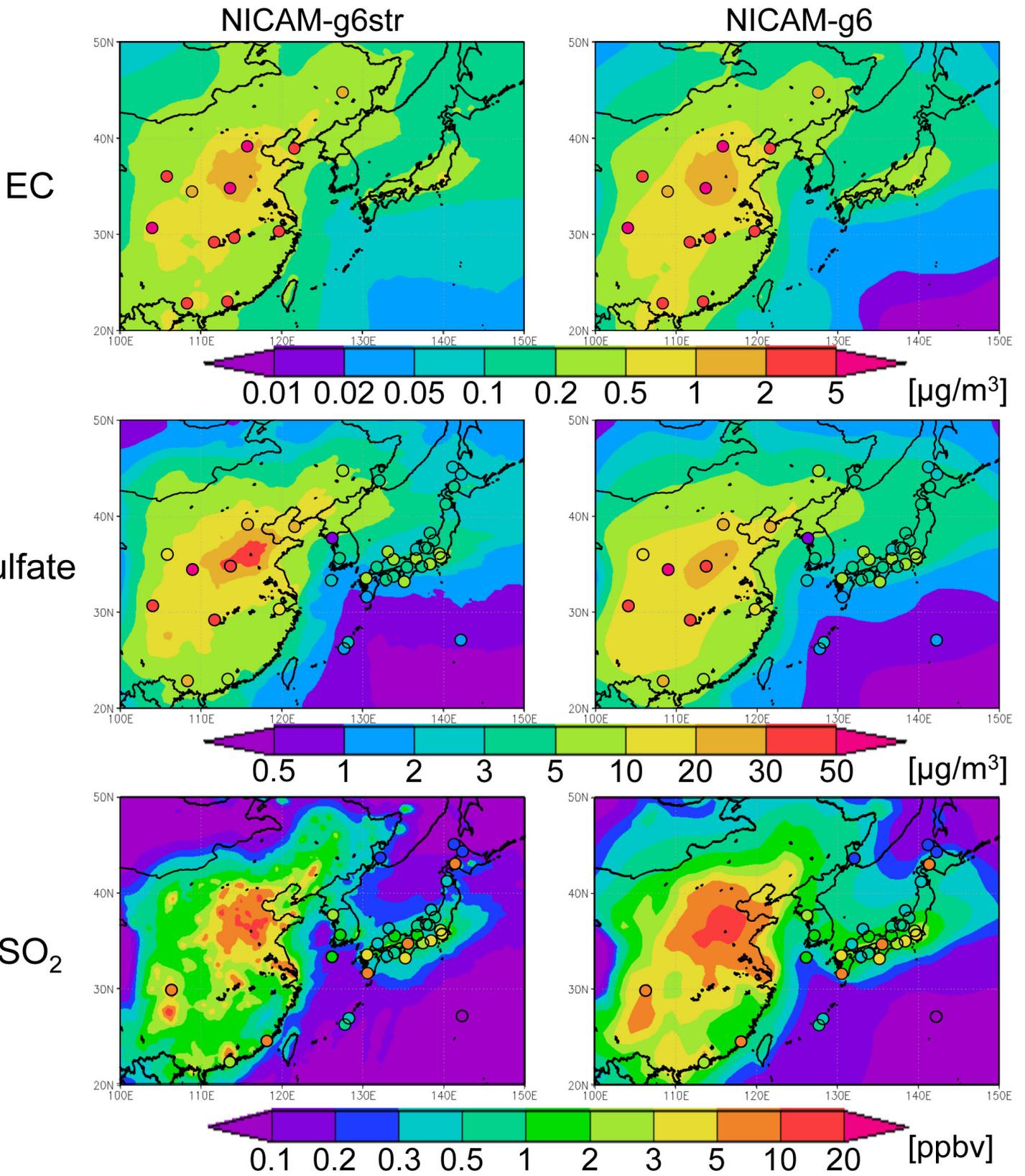
(a) Maebashi [139.10°E, 36.40°N]



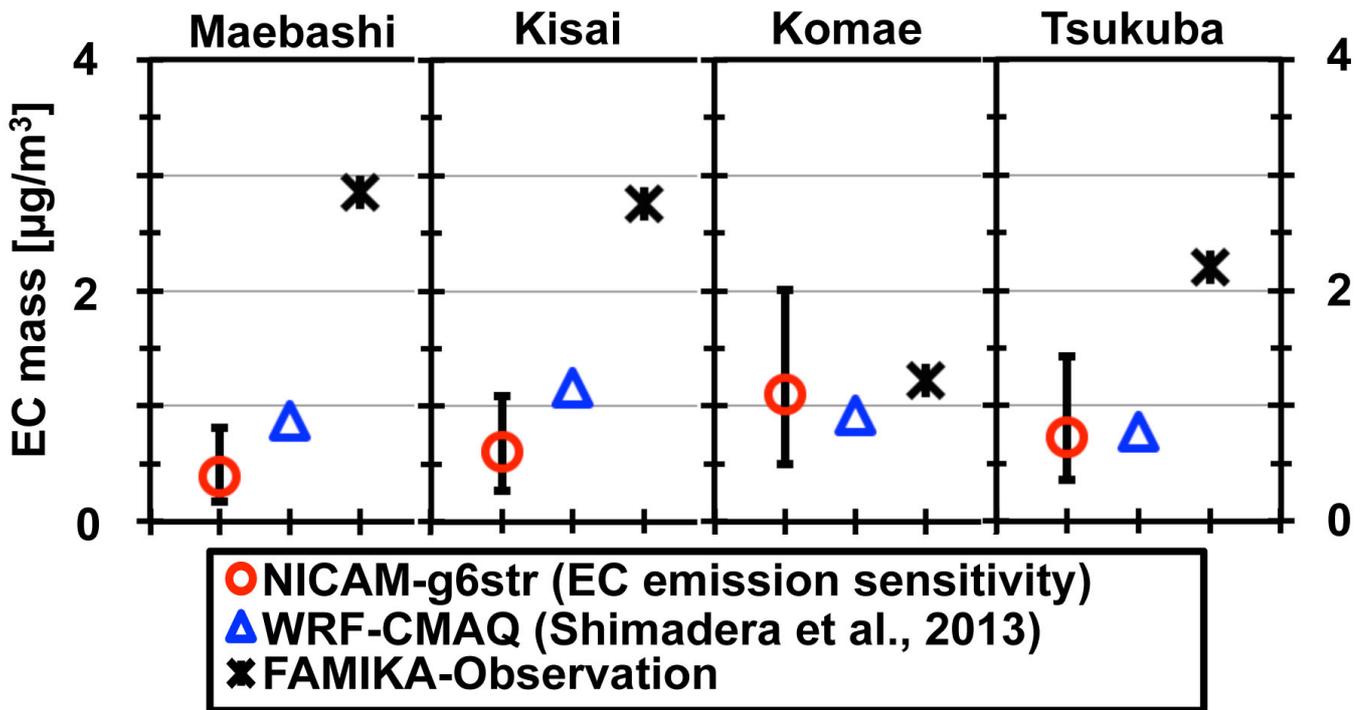
(c) Komae [139.58°E, 35.64°N]







(a) EC mass concentration at FAMIKA sites



(b) Sulfate mass concentration at FAMIKA sites

