We thank the referee #1 for the comments and suggestions. The Point by Point Clarifications to the comments and suggestions are as follows;

Response to comments of anonymous referee #1

[C1-1] This paper describes application of Stretch-NICAM-SPRINTERS to Kanto region in Japan. The stretched-grid system embedded in this models realizes more efficient simulations over target regions in finer resolutions. In addition, it is superior to general regional models because it does not need to apply a nesting technique and boundary conditions.

It appears that Stretch-NICAM-SPRINTERS has a great possibilities in its concept. However, I do not have any impressions only from this paper that this model has a good performance. It is too ambiguous how the authors have judged that this model was capable of simulating meteorological fields and anthropogenic primary and secondary particles. If the authors judge so, the concrete criteria should be shown.

[A1-1] Thank you very much for reading our manuscript and giving us useful comments for improving the manuscript. In this study, the main purpose is to show the model performance of simulating aerosols with the stretched-grid system. Basically, when we judged that our presented model has a good performance, the model captured important features (e.g., daily and weekly variations of the meteorological and aerosol fields, their magnitudes in daily, weekly, and monthly averages etc.) over the target regions and the simulated results were within ranges obtained by general regional aerosol-transport models, e.g., WRF-CMAQ. To show these results, we have compared Stretch-NICAM-SPRINTARS (NICAM-g6str in the revised manuscript) with other models, i.e., Global-NICAM-SPRINTARS or NICAM-g6 in the revised manuscript and sometimes WRF-CMAQ, as you suggested in [C1-4]. In the evaluation of the meteorological fields, we have showed the large-scale circulations of basic physical variables over East Asia (Figure 3 of the revised manuscript, suggested by the referee #2 on [C2-5]). The results indicate that the NICAM-g6str-simulated temperature and winds are consistent to the NICAM-g6-simulated and the NCEP-FNL-reanalyzed ones. We also compared the meteorological fields (temperature, RH, and winds) and precipitation in Figures 4-10 of the revised manuscript and Table 1. We have modified our comments on the analysis to clarify them. As for EC, sulfate, and SO₂, we have modified the figures and added a new table to the revised manuscript to show the statistical parameters with the results of WRF-CMAQ by Shimadera et al. (2013). PM2.5 has been evaluated using both NICAM-g6str and NICAM-g6 simulations. Please read the details in the revised manuscript.

[C1-2] The stretched-grid system should be one of advantages of Stretch-NICAM-SPRINTERS. On the other hand, the treatment of aerosols is much more simplified than general regional models (no nitrate, ammonium in a fixed ratio, prescribed oxidants, etc.). This paper does not describe that a good performance (which the authors judged) was obtained from the former or the latter. Are the authors indicating that the simplified treatment of aerosol enough to represent aerosol over Kanto region?

[A1-2] In the revised manuscript, the comparison between NICAM-g6str and NICAM-g6 indicates that the meteorological and aerosol fields obtained by NICAM-g6str are better than those obtained by NICAM-g6. It suggests that the higher horizontal resolution under the stretched grid system leads to a better performance. After the comparisons shown in the revised manuscript, we conclude that even the simplified aerosol module is applicable for the regional simulation if the module is coupled to dynamic cores with high horizontal resolution. We have added this point to summary in the revised manuscript, as you suggested; (1) no nitrate and ammonium in a fixed ratio and (2) prescribed oxidants. The point (1) is addressed in [A1-10]. The point (2) is described below.

Prescribed oxidant for the use of sulfate formation is not probably crucial for predicting weekly- and monthly-averaged sulfate mass concentrations only by taking into account for diurnal and seasonal variations of the prescribed oxidant (which is based on our experiments). The statement is consistent with the results of the averaged sulfate mass concentration obtained by our model (NICAM-g6str) are comparable to those obtained by a regional aerosol-transport model, WRF-CMAQ. However, our model sometimes misses hourly variations obtained by WRF-CMAQ. Therefore, at least we can say that it may be important to predict hourly variations of the sulfate formation, especially during the daytime, because the oxidants largely depend on solar downward surface radiation and indirectly on clouds. We have added these comments on the revised manuscript.

[C1-3] In addition, the authors describe Stretch-NICAM-SPRINTERS are potentially superior in simulations of transboundary air pollution. However, any discussions of transboundary air pollution are missing.

[A1-3] Thank you very much for your comments. To show any evidence to support our statement, we need to compare the results obtained by Stretch-NICAM with those obtained by general regional models. Strictly speaking, we need to prepare both a regional model with the same platform as the Stretch-NICAM and the output of Stretch-NICAM as the lateral boundary conditions. Although we mentioned the detail in our answer ([A1-6] to [A1-8]), we did not show clear evidence for the superiority of our presented model to general regional models, in terms of transboundary air pollution. Therefore, we have deleted the sentence (L.2 of P.135 in the manuscript).

[C1-4] I do not think that the assessment of the public health impact is suitable to be included in the current form in this paper. I suppose the main objective of this paper is to show advantages of Stretch-NICAM-SPRINTERS. If the authors are willing to include this part, the advantages of Stretch-NICAM-SPRINTERS in the results should be clearly stated for example by comparing results with those obtained by other models (e.g. MIROC-AOGCM). The current results may cause confusion because mortality in 2030 would increase whereas PM2.5 concentration decreases. It is due to changes in the age distribution. In this case, I think it is necessary to describe how to predict future population and its distribution in ages in details. However, explanations on the population data used in this study are almost missing in the current manuscript. I also

think such a discussion would be a topic to be described in a separate paper focusing on the assessment of the public health impact.

[A1-4] Thank you very much for your comments and suggestions to splitting our manuscript into two. Yes, we agree to your idea and we have rearranged our manuscript to mainly focus on model results of Stretch-NICAM-SPRINTARS without topics of health impact and scenario experiments.

In addition, as you suggested, we have compared Stretch-NICAM-SPRINTARS (NICAM-g6str in the revised manuscript) with Global-NICAM-SPRINTARS (NICAM-g6 in the revised manuscript). Detail is found in the revised manuscript. Although you suggested a use of MIROC-AOGCM as a reference, differences in the dynamic and physics parts between MIROC and NICAM are not small, so it is difficult to properly evaluate our presented model (Stretch-NICAM-SPRINTARS). Therefore, we have selected the model, Global-NICAM-SPRINTARS, which has the identical dynamic and physics modules to Stretch-NICAM-SPRINTARS, although the horizontal resolution were different due to the insufficient computer time to integrate Global-NICAM-SPRINTARS with the finer resolution.

[C1-5] Most of figures are too obscure to recognize if the description in the main text is valid. Especially, the described features in the horizontal distribution over Kanto regions are hard to be recognized in contour figures.

[A1-5] Apologies for using the rainbow color without clear borderlines in figures. According to your suggestions, we have re-plotted all of figures using clear color bars.

Specific comments:

[C1-6] Line 1 in Page 135

How is Stretch-NICAM-SPRINTERS potentially superior to general regional models? [C1-7] Indeed, a nesting technique or boundary conditions are necessary in regional models. Does it mean that a nesting technique or boundary conditions have any problems to represent transboundary air pollution accurately? Are there any references which imply such problems? Or, is it just complicated to apply a nesting technique or boundary conditions? Is it appropriate to determine that Stretch-NICAM-SPRINTERS is "potentially superior" only by this reason?

[C1-8] Line 25 in Page 136

Again, it is not clear that how the stretched-grid is more suitable for the current study compared with general regional models.

[A1-6]&[A1-7]&[A1-8] Thank you very much for your comments. As you suggested, we need to add any evidence or references, which show problems of simulating transboundary air pollution using the general regional models. We have reconsidered this issue. One reference (Bhaskaran et al., 1998) pointed out the critical problem to simulate precipitation patterns related to intra-seasonal variability such as monsoon using the regional model through a dynamic downscaling technique. However, as far as we know, such problems for simulating the air pollution have not yet been reported.

Although the lateral boundary conditions originally cause noises to perturb the airflow inside the domain, researchers have paid attention to minimize such noises by using various techniques (e.g., Nakanishi, 2001). So far, if users choose specific techniques and/or proper lateral boundary conditions, they may often escape problems to accurately simulate the transboundary air pollution. In contrast, our proposed method (Stretch-NICAM) is not required to consider such complex issues. In terms of the nesting technique and boundary conditions, we can safely say that Stretch-NICAM is absolutely superior to general regional models. In addition, as we mentioned in the manuscript, the model framework of the Stretch-NICAM-SPRINTARS is identical to that of the Global-NICAM-SPRINTARS without special modifications. This point is also very important for modelers to sophisticate the module such as aerosol module, because the model frameworks of regional models are usually different from those of global models, which often prevent expanding regional simulations to global simulations. This is based on the assumption that aerosol modules implemented to regional models are generally more detailed than those implemented to global models, because the spatial and temporal scales of the main target in the regional simulations are finer than those in the global simulations. Recently, owing to increases in the computational resources, global models have started to focus on the finer scales (Suzuki et al., 2008). To further develop the aerosol modules in the global models, the aerosol modules implemented to the regional models should be reflected to the global models, even though the implementation may be complex or perhaps troublesome. In the present study, since we cannot show clear evidence of the superiority to general regional models, we have removed the sentence (Line 1, P.135 in the manuscript) from the revised manuscript. However, we can safely say that our presented model can be applicable for simulating regional aerosols.

[Reference]

Bhaskaran, B., Murphy, J. M., and Jones, R. G.: Intraseasonal oscillation in the Indian summer monsoon simulated by global and nested regional climate models, Monthly Weather Review, 126, 3214-3134, 1998.

Nakanishi, M.: A Lateral boundary condition suitable for the one-way nesting scheme, Tenki, 49(2), 117-128, 2001, in Japanese

[C1-9] Line 4 in Page 138

Anthropogenic SOAs from toluene and xylene are disregarded in this study. However, Morino et al. (2010c) implied that anthropogenic SOAs are important during FAMIKA. Potential influences on simulated PM2.5 should be discussed.

[A1-9]

Thank you very much for your suggestions. We have added this point to the paragraph of PM2.5 (section 3.2.3) in the revised manuscript; "At all sites, the possible underestimation of SOA may be a critical issue, as shown in the fact that the clear diurnal variation of PM2.5 during August 4-9 and suggested by previous studies (Matsui et al., 2009; Morino et al., 2010c). Morino et al. (2010c) implied that over the Kanto area SOA from anthropogenic sources, which were disregarded in this study, are large portion of total carbonaceous aerosols, even though WRF-CMAQ does not

correctly reproduce such carbonaceous aerosols. More sophisticated SOA module, e.g., volatility basis-set approach proposed by Donahue et al. (2006) based on the categorization of organic vapors with similar volatility, is required for to produce SOA with higher accuracy."

[C1-10] Line 19 in Page 138

According to Morino et al. (2010b), 1-3 micrograms per cubic meters of nitrate were observed at FAMIKA. This magnitude is comparable to or even more than EC. Although nitrate is not abundant in summer, just disregarding nitrate is too rough.

[A1-10]

Thank you very much for your comments. Surely, the nitrate mass concentration is comparable to the EC mass concentration, even in summer of Kanto area, although the nitrate mass concentration in summer tends to be lower than the sulfate one. As you pointed, disregarding nitrate may be too rough. In this study, however, we would like to show whether our presented model could be applied to a regional simulation as a general aerosol-transport model. Since the stretched-grid system in this study was used in the previous study for simulating tropical cyclones and tropical convective clouds over oceans (e.g., Satoh et al., 2010; Arakane et al., 2013), it was not adequately evaluated over the target region focused in this study (megacities over mid-latitudes). Therefore, for this purpose, we have compared representative primary and secondary aerosols in summer of Japan. We chose sulfate as a representative secondary aerosol. The global and regional modelings for sulfate, which is formed from SO₂ in the atmosphere, are more deeply understood compared to modelings for the other secondary aerosols such as nitrate and organic aerosols (e.g., Barrie et al., 2001; Holloway et al., 2008; Hallquist et al., 2009; Morino et al., 2010a, 2010b). In addition, sulfate is the largest contributor to the total secondary inorganic aerosols (e.g., Zhang et al., 2007), and the sulfate mass concentrations are larger than that the nitrate ones in August 2007 over the Kanto area (Morino et al., 2010c). Furthermore, in summer over Japan and East Asia, the difference in the mass concentrations between sulfate and nitrate is higher than that in winter. Therefore, we disregard nitrate in this study. We have added them to section 1 of the revised manuscript and modified section 2.2 as follows; "The nitrate in this study is disregarded, primarily because the main objective in this study is modeling of sulfate as a representative secondary aerosols and secondly because the nitrate mass concentrations are lower than the sulfate ones with the target of August 2007 in Japan (Morino et al., 2010c)".

[References]

Barrie, L. A., YI, Y., Leaitch, W. R., Lohmann, U., Kasibhatla, P., Roelofs, G.-J., Wilson, J., McGovern, F., Benkovitz, C., Melieres, M. A., Law, K., Prospero, J., Kritz, M., Bergmann, D., Bridgeman, C., Chin, M., Christensen, J., Easter, R., Feichter, J., Land, C., Jeuken, A., Kjellstrom, E., Koch, D., and Rasch, P.: A comparison of

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large-scale atmospheric sulphate aerosol models (COSAM): overview and highlights, Tellus, 53B, 615-645, 2001.

Holloway, T., Sakurai, T., Han, Z., Ehlers, S., Spak, S.N., Horowitz, L. W., Carmichael, G. R., Streets, D. G., Hozumi, Y., Ueda, H., Park, S. U., Fung, C., Kajino, M., Thongboonchoo, N., Engardt, M., Bennet, C., Hayami, H., Sartelet, K., Wang, Z., Matsuda, K., and Amann, M.: MICS-Asia II: Impact of global emissions on regional aiq quality in Asia, Atmos. Environ., 42, 3543-3561, 2008.

[C1-11] Line 11 in Page 141

Stretch-NICAM-SPRINTERS cannot be used for a long-term simulation. However, the sentence in the line 4 in the page 134 says that the stretch grid overcomes the limitation (requirement of vast computer resources for highly resolved calculations). What a temporal scale is expected to apply Stretch-NICAM-SPRINTERS?

[A1-11]

We intended to mention that "long-term simulation" in P.141 of the manuscript is "several thousands integration as an atmosphere-ocean coupled model", whereas the "long-term simulation" in P.134 of the manuscript means "several decades integration as an atmospheric model". So, we have modified the latter point (by adding "for decades" to section 1 of the revised manuscript). We have removed the former point from the revised manuscript.

[C1-12] Line 24 in Page 141

What is the horizontal resolution of MIROC-CHASER?

[A1-12] Actually this part has been removed from the manuscript, because we have removed the results of scenario experiments. In the standard experiment, we also used prescribed oxidant distribution from MIROC-CHASER with the spatial resolution of 2.8 degree and the temporal resolution of three-hourly averaged monthly averages. We have added the explanation to the section 2.3 of the revised manuscript.

[C1-13] Line 26 in Page 143

How can be judged that poor performance at Maebashi and Kisai is due to the topography? What is the detailed configuration of the topography data used in this study?

[A1-13]

Thank you very much for your comments. First, the source of the topography data used in this study and shown in Figure 1 is GTOPO30. So we have added the following comments to the revised manuscript (section 2.1 and the caption of Figure 1); "the topography used in this study is based on GTOPO30 (the horizontal resolution is 30 arc seconds, that is approximately 1 km) courtesy of the U.S. Geological Survey." We have changed the grey contour in Figure 1 to clarify the values. Second, we need to add some explanation of how the topography does influence the meteorological fields at Maebashi (139.10°E, 36.40°N) and Kisai (139.56°E, 36.09°N). In Figure 6 of the revised manuscript, you can find that especially during the daytime at Maebashi and Kisai, the NICAM-g6str-simulated winds are not close to the observations, which show southeasterly winds (that is long sea breeze toward Maebashi Plateau surrounded on three sides by mountains around Maebashi). The observed winds are caused by daytime meso-scale thermal lows developed over the central Japan covering the Japanese Alps (Kuwagata and Sumioka, 1991). The Japanese Alps with the highest terrain in Japan can affect the local meteorological fields even around 100-200 km away (Kitada et al., 1998). Therefore, we can conclude that the horizontal resolution in this study using Stretch-NICAM cannot fully resolve the complex terrains of the Japanese Alps and the Maebashi plateau. These comments had been inserted to section 3.1 of the revised manuscript; "It suggests that it is inadequate to simulate the wind patterns and diurnal transitions near high mountains around the Kanto region, whereas it is adequate to simulate them around the center of Kanto region."

[Reference]

Kuwagata, T., and Sumioka, M.: The daytime PBL heating process over complex terrain in central Japan under fair and calm weather conditions, Part III: Daytime thermal low and nocturnal thermal high, J. Met. Soc. Japan, 69(1), 91-104, 1991 Kitada, T., Okamura, K., and Tanaka, S.: Effects of topography and urbanization on local winds and thermal environment in the Nohbi Plain, coastal region of central Japan: A numerical analysis by mesoscale meteorological model with a k-e turbulence model, J. Applied Met., 37, 1026-1046, 1998.

[C1-14] Line 12 in Page 144

It is very difficult to recognize the overestimation of the precipitation in the Sea of Japan, Kyusyu, and the main island of Japan in Fig. 9.

[A1-14] Apologies for the unclear color maps. In the revised manuscript, we recalculated NICAM-g6str using longer spinup time (one and a half months) than that in the previous manuscript. As a result, the precipitation distribution was somewhat different from that obtained by the previous simulation. In the revised manuscript, we have added the following comments to section 3.1; "During early August 2007, mainly due to passing of a typhoon over the western Japan, Okinawa, and Korea, the August mean precipitation in the western Japan is larger than that in the eastern Japan, especially the Kanto region (Figure 8 of the revised manuscript). The monthly mean precipitation is estimated to be more than 200 mm/month over the western Japan, whereas that is estimated to be less than 50 mm/month over the eastern Japan. In the NICAM-g6str results, the horizontal distribution of the August mean precipitation in the East China Sea, the Sea of Japan near the Japan coast, and Korea are closer to those derived from MSM and GSMaP than those by NICAM-g6. In the Kanto area, however, the NICAM-g6str-simulated precipitation with ranges of 50-200 mm/month is overestimated compared to the MSM and GSMaP results. The NICAM-g6-simulated precipitation over the Kanto area is also much overestimated, with ranges of 100-200 mm/month."

[C1-15] Line 20 in Page 145

The overestimation of the simulated precipitation shown in Fig. 9 may cause the underestimation of the simulated sulfate concentrations at Hedo. However, the sentences in the line 14 in the page 144 says that all results generally shows similar patterns of the occurrence of heavy precipitation in the East China Sea especially near Okinawa in which Hedo is located. They may cause confusions.

[A1-15]

Thank you for your suggestions. Figure 8 of the revised manuscript shows that the heavy precipitation area with more than 500 mm/month is found in the East China Sea especially near Okinawa by both the observations (MSM and GsMAP) and simulations (NICAM-g6str). However, when the temporal variation of the NICAM-g6str-simulated precipitation and the predictive value of daily precipitation using NICAM-g6str are compared with those obtained by the observations in Figures 9 and 10 of the revised manuscript, at Okinawa (Panel (u) of Figure 9 in the revised manuscript) the timing of the NICAM-g6str-simulated precipitation is different from that obtained by the observation and the predictive value using NICAM-g6str is the lowest among all of the sites. Therefore, the underestimation of the NICAM-g6str-simulated sulfate at Hedo of Okinawa may be caused by the possible underestimation of the sulfate from transboundary air pollution, which might be caused by large uncertainty of the simulated precipitation related to the typhoon, as the referee #2 pointed out in C2-3. We have modified this part section 3.2.1 of the revised manuscript.

[C1-16] Line 4 in Page 147

Why do the offline oxidants not alter sulfate concentrations so much?

[A1-16] Thank you very much for your comments. In this sensitivity tests for oxidants, the SO₂ oxidation by OH radical strongly depends on the OH concentrations as well as the cloud cover area, whereas the SO₂ oxidation by ozone and hydrogen peroxide mainly depends on their concentrations, the cloud cover area, and the cloud water content. The cloud distributions are modulated by some feedbacks of the sulfate formation through the aerosol direct and indirect effects. These various pathways can determine the magnitude of the sulfate formation. As a result, the sensitivity of the OH radical concentrations to the simulated sulfate concentration is smaller than that we expected and that to the SO₂ emissions, as shown in Figure 16(b) of the revised manuscript. We have added these comments to section 3.2.2 of the revised manuscript.

[C1-17] Line 11 in Page 147

Why is PM2.5 included here? It is also one of the validations of Stretch-NICAM-SPRINTERS described in the section 3, isn't it?

[A1-17] As you suggested, we have modified this part (PM2.5) to section 3.2.3.

[C1-18] Line 10 in Page 148

Indeed, when the results of PM2.5 obtained by Stretch-NICAM-SPRINTERS are used in an estimation of health impacts due to PM2.5, the bias should be minimized. However, Stretch-NICAM-SPRINTERS has been immediately applied to estimate health impacts in the subsequent subsection without minimizing the bias. That is obviously inconsistent.

[A1-18] We actually did a bias correction by multiplying twice PM2.5 values for the application. However, we have deleted the application issue from the revised manuscript.

[C1-19] Line 19 in Page 148

It is very difficult to recognize that sulfate mass concentrations over the Kanto region decrease from the present to 2030 in Fig. 18 (and Fig. 14?).

[A1-19] Apologies for using unclear color maps. We have corrected all of figures.

[C1-20] Line 25 in Page 148

Why is the largest sulfate mass concentration in Ibaraki unrealistic? As shown in Fig. 11, the highest observed sulfate concentration is at Tsukuba, which is certainly located in Ibaraki. It is not strange that concentrations of secondary components are higher in downwind regions than source regions. It is very surprising that large differences among prefectures are found in MIROC with coarser resolutions while differences among prefectures are very small in NICAM in sulfate concentrations shown in Fig. 19. Are there any reasons?

[A1-20] Thank you very much for your comments, but we have removed this part from the revised manuscript. The strong peak in the coarse resolution causes the higher variability in the sulfate concentrations.

[C1-21] Fig. 6(d) Ayase -> Adachi

[A1-21] Thank you for your correction.

We thank the referee #2 for the comments and suggestions. The Point by Point Clarifications to the comments and suggestions are as follows;

Response to comments of anonymous referee #2

[C2-1] A global non-hydrostatic model with a stretched grid system is used to simulate aerosol distributions around the highly populated Kanto region of Japan during the month of August 2007. The stretched grid system uses a fine mesh (allows high resolution) over the target region increasing to larger mesh (lower resolution) on the opposite side of the globe. This type of grid appears very promising as it eliminates the need for nesting techniques and boundary conditions required in regional air quality models. Simulated meteorological and aerosol variables are evaluated against a range of ground-based measurements and the application of this modelling system for air quality forecasting is advocated. The model is then run in a future climate scenario set-up to assess the impact of future aerosol emissions on mortality in Japan.

The quality and content of this manuscript needs to be greatly improved before publication should be considered. The results and conclusions drawn in the first part of the manuscript in which the Stretch-NICAM-SPRINTARS is run and evaluated for August 2007 are in my opinion inconsistent. The authors conclude that the "simulations of Stretch-NICAM-SPRINTARS are generally successful in simulating the air pollution over Japan and are adequate as a new regional model for simulations over the Kanto region". However, there are clear shortcomings in the current simulations. Omission of nitrate aerosol, simplified sulphur and SOA chemistry are major barriers to a skillful air quality forecast.

[A2-1] Thank you very much for reading our manuscript and giving us useful comments for improving the manuscript. In this study, the main purpose is to show the model performance of simulating aerosols with the stretched-grid system. For this purpose, we have shown that our presented model captures important features (e.g., diurnal and weekly variations of the meteorological and aerosol fields, their magnitudes in daily, weekly, and monthly averages etc.) over the target regions and the simulated results were within ranges obtained by general regional aerosol-transport models, e.g., WRF-CMAQ. Since the stretched-grid system in this study was used in the previous study for simulating tropical cyclones and tropical convective clouds over oceans (e.g., Satoh et al., 2010; Arakane et al., 2013), it was not adequately evaluated over the target region focused in this study (megacities over mid-latitudes).

Therefore, for this purpose, we have compared representative primary and secondary aerosols in summer of Japan. We chose sulfate as a representative secondary aerosol. The global and regional modelings for sulfate, which is formed from SO_2 in the atmosphere, are more deeply understood compared to modelings for the other secondary aerosols such as nitrate and organic aerosols (e.g., Barrie et al., 2001; Holloway et al., 2008; Hallquist et al., 2009; Morino et al., 2010a, 2010b). In addition, sulfate is the largest contributor to the total secondary inorganic aerosols (e.g., Zhang et al., 2007), and the sulfate mass concentrations are larger than that the nitrate ones in August 2007 over the Kanto area (Morino et al., 2010c). Furthermore, in summer over Japan and East Asia, the difference in the mass concentrations between sulfate and nitrate is higher than

that in winter. Therefore, we disregard nitrate in this study. We have added them to section 1 of the revised manuscript.

Surely, the nitrate aerosol is also important to precisely forecast the air quality. Many air quality models (e.g., CMAQ) have tried to simulate nitrate aerosols to get closer results to the measurements. However, the variability of the model prediction for the nitrate aerosols even among general regional aerosol-transport models seems to be very large (e.g., Morino et al., 2010a). The one of the reasons of the gap between the simulation and observation is uncertainty of the thermodynamical module, which is implemented to host models. The second possible reason is high uncertainties of emission inventory for ammonia (e.g., Shimadera et al., 2014). Under the current situation, we feel it is very difficult to adequately validate nitrate aerosols using our proposed model. The nitrate simulation using our present model is the next work for winter and future scenarios and this shortcoming of our present model has been mentioned in summary of the revised manuscript.

As for secondary organic aerosols (SOA), our model is required to improve the simplified SOA chemistry and implement SOA from anthropogenic sources. However, as we mentioned in the nitrate part, the primary purpose of this study is to confirm that the stretched model can be an aerosol transport model to predict the concentrations over the Kanto area. In addition, as you know, the SOA chemistry includes a large uncertainty (underestimation) of their prediction (e.g., Hallquist et al., 2009; Matsui et al., 2009; Morino et al., 2010c). Although SOA become the most important pollutants over East Asia and its modeling have been developed by many attempts such as volatility basis-set approach proposed by Donahue et al. (2006) based on the categorization of organic vapors with similar volatility, their implementation to our model is beyond the present study. The shortcoming of the simplified SOA chemistry in this study has been mentioned in the comparison of PM2.5 at Japanese sites (section 3.2.3 of the revised manuscript).

As for sulfur chemistry, we mainly simplified two points; (1) prescribed oxidants and (2) fixed sizes of the sulfate. Although the point (2) is crucial for predicting sulfate size distribution, ignorance of the point (2) can be accepted for predicting sulfate mass concentrations. Reversely speaking, treating of the size distribution does not always improve the simulated sulfate mass concentrations, because the degree of freedom increases (e.g., Kajino and Kondo, 2011). In contrast, the point (2) is not probably crucial for predicting weekly- and monthly-averaged sulfate mass concentrations only by taking into account for diurnal and seasonal variations of the prescribed oxidant (which is based on our experiments). The statement is consistent with the results of the averaged sulfate concentration obtained mass by our model (Stretch-NICAM-SPRINTARS) are comparable to those obtained by a regional aerosol-transport model, WRF-CMAQ. However, our model sometimes misses hourly variations obtained by WRF-CMAQ. Therefore, at least we can say that it will be important to predict hourly variations of the sulfate formation, especially during the daytime, because the oxidants largely depend on solar downward surface radiation and indirectly on clouds. We have added these comments to section 3.2.1 and 3.2.2 of the revised manuscript.

[References]

Arakane, S., Satoh, M., and Yanase, W.: Excitation of deep convection to the north of tropical storm Bebinca (2006), J. Meteorol. Soc. Japan, 92(2), 141-161, doi:10.2151/jmsj.2014-201, 2014.

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[C2-2] Indeed Figures 10-12 highlight the clear underestimation in aerosol fields and the model clearly misses a number of peak SO2 and PM episodes.

[A2-2] Thank you very much for your comments. In the revised manuscript, we have compared our presented model (Stretch-NICAM-SPRINTARS, which has been renamed as NICAM-g6str), with other models (Global-NICAM-SPRINTARS named as NICAM-g6) and WRF-CMAQ shown by Shimadera et al., 2013). As the referee #1 suggested in [C1-4], we have shown other model with the same dynamic core (NICAM), although the horizontal resolution were different due to the insufficient computer time to integrate Global-NICAM-SPRINTARS with the finer resolution. As you suggested, our model (both NICAM-g6str and NICAM-g6) does not always capture the observed peaks. However, WRF-CMAQ also sometimes misses the observed peaks of EC and sulfate. The results obtained by NICAM-g6str are within ranges obtained by WRF-CMAQ. As for SO₂, we have modified the figure to plot hourly-averaged SO₂ concentrations and added more analysis to section 3.2.1 of the revised manuscript. In

the previous studies, the comparison of SO_2 between the simulation and observation was extremely limited, with the exception of in Figure 4 of Morino et al. (2010b), which showed large differences in the SO_2 concentrations between WRF-CMAQ and the observation by more than a factor of two. The R between the NICAM-g6str-simulations and the observations are low, with the exception of Komae where the R value is 0.62 (safely acceptable), but are approximately within ranges obtained by WRF-CMAQ in Morino et al. (2010b). Therefore, we have judged that our presented model (NICAM-g6str) is capable of simulating the aerosol species.

[C2-3] Poor performance in precipitation fields will seriously affect the aerosol transport within the simulations in particular the impact of trans-boundary pollution from China.

[A2-3] As you mentioned, the performance in the precipitation fields had a large impact on the aerosol distribution. In August, North Pacific High (or Ogasawasa High) mainly brings clear weather around Japan. A frequency of the precipitation is usually limited, but the total amount of the monthly mean precipitation is not small, because of typhoons and shower rain. The prediction of the typhoon and the precipitation amount by the typhoon is generally difficult. In addition, usual numerical modeling has still difficulties in predicting heavy rainfalls induced by orographic or synoptic forcing and small-scale convective rainfalls (e.g., Kawabata et al., 2011).

During the early August 2007, mainly due to passing of a typhoon over the western Japan, Okinawa, and Korea, the August mean precipitation in the western Japan is larger than that in the eastern Japan, especially the Kanto area. The monthly mean precipitation is estimated to be more than 200 mm/month over the western Japan, whereas that is estimated to be less than 50 mm/month over the eastern Japan. The NICAM-g6-simulated precipitation over the Kanto area with the range of 100-200 mm/month is also much overestimated.

Although the total amount of the precipitation obtained by NICAM-g6str is overestimated, the frequency of the precipitation obtained by NICAM-g6str is close to that obtained by the in-situ measurements of AMeDAS (Figure 9 of the revised manuscript). In Figure 9 of the revised manuscript, which shows the temporal variations in the amount of precipitation per day at 21 Japanese sites, the observed precipitation is extremely limited during August 7-19 in the Kanto region. In other regions, the magnitude of the precipitation is strong, although the precipitation is sporadic. Figure 10 of the revised manuscript illustrates the predictive value of daily precipitation, defined as the ratio of the number of days where the model correctly predicts the weather (less than 1 mm/day or more than 1 mm/day) to the number of the whole days. In the NICAM-g6str results, the predictive values at most of sites over the Kanto region and four sites over non-Kanto region such as Nagoya and Osaka are calculated to be more than 85%. The predictive values obtained by NICAM-g6str are mostly higher than those estimated by NICAM-g6.

During the rainy days such as August 20, 22 and 23 over the Kanto region, both NICAM-g6str and NICAM-g6 capture the precipitation, whereas NICAM-g6str reproduces greater amounts of the precipitation and NICAM-g6 reproduces longer

periods and larger areas compared to the observations. Even NICAM-g6str does not always capture a sudden shower, as general meteorological models have difficulties in properly simulating this type of precipitation system, as mentioned in the first paragraph.

Therefore, these larger uncertainties of the predicted precipitation can cause the large uncertainties of the predicted transboundary pollution from China to Japan and Kanto area. Although we can say that the precipitation fields simulated by NICAM-g6str are not so bad, but we have added the following comments to the revised manuscript; "The underestimation of both NICAM-g6str and NICAM-g6 simulated sulfate concentrations is caused by a possible underestimation of transboundary sulfate from the continent, which is attributed to a large uncertainty of the precipitation fields modulated by typhoon in the early August."

Kawabata, T., Kuroda, T., Seko, H., and Saito, K.: A Cloud-Resolving 4DVAR Assimilation Experiment for a Local Heavy Rainfall Event in the Tokyo Metropolitan Area. Mon. Wea. Rev., 139, 1911–1931, 2011.

[C2-4] While the authors highlight various developments/improvements which should be conducted in future work to improve the quality of these simulations these points/the limitations of the current simulations should also be emphasized when discussing the results in Section 3.2.

[A2-4] Thank you very much for your comments. We have added these points to each section.

[C2-5] There is no mention of the global performance of the model. Is it capable of producing the large scale circulations required for an adequate simulation over the target region? Perhaps an evaluation of large-scale circulations against reanalysis could be performed. I am surprised given that the model is nudged that there is such discrepancies in the circulation. 2D spatial plots of the circulation compared with reanalysis or satellite observations would give a nice depiction of the models ability in capturing the general flow.

[A2-5] Thank you very much for your suggestions. In section 3.1 of the revised manuscript, we have added the results of circulation over Asia region (100°E-170°E, 10°N-50°N) and have confirmed NICAM-g6str as well as NICAM-g6 are capable of simulating the large-scale circulation over Japan. Please see the detail in section 3.1 of the revised manuscript.

[C2-6] From the current evaluation it is not clear whether the simulations using the stretched grid model are superior to a more conventional nested uniform grid regional model. An evaluation against a regional model would put the current study in much better context. Furthermore the authors claim that the computational cost of running the stretched model is 256 times smaller than a global model with a uniform grid of the

same high resolution as in the target region. Given the application to regional air quality the authors should really be comparing the cost to a regional model over the same target domain as used in this study.

[A2-6] Thank you very much for giving us the important comments. Surely, we often mentioned the advantage of the stretched-grid model proposed in this study, compared to a more conventional nested uniform grid regional model. However, we did not show clear evidence of the advantage.

As you suggested that an evaluation against a regional model would put the current study in much better context, we just have added a regional aerosol-transport model, WRF-CMAQ, shown in Shimadera et al. (2013) in the comparison of EC and sulfate at FAMIKA site, because it is very hard for us to execute different regional models with the same experimental conditions under this study. Since we cannot show clear evidence of the superiority to general regional models, we have removed the sentence (Line 1, P.135 in the manuscript) from the revised manuscript. However, we can safely say that our presented model can be applicable for simulating regional aerosols.

The second point you suggested is also very important, because model users often worry about the computational cost using models. Your suggestion is proper request and we would like to answer it truly, but it is extremely difficult to compare the cost of our model with other regional model. As far as we know, there are no studies for comparison in the computational cost among different regional air quality models. The reason is mainly caused by the difficulty in setting the same platform including the dynamic core and physical processes. The comparison in the whole model is rather difficult than that in each specific module. Now a Team NICAM is developing a regional model coupled to NICAM (we call it Diamond-NICAM, because a diamond (two triangles) panel used in the regional simulation cuts off the regular icosahedron). Ideally, the computational cost of Diamond-NICAM is smaller than that of Global-NICAM (NOT Stretch-NICAM!) by ten times, but we have never estimated a difference in the computational cost between Stretched-NICAM and Diamond-NICAM. This will be conducted near the future. However, when we compare the computational cost in Stretch-NICAM with that in different regional models, we need to take into account for the time to prepare lateral boundary conditions, the number of the nesting, and the domain area (actually the number of the grid). The various selections of the experimental conditions prevent estimating the actual required time to calculate them. Theoretically, we can safely say that it may take more time to calculate the air pollution using the stretched-grid system than conventional regional models over the target region, because the stretched-grid system requires the calculation outside the target domain (that means the grid number in the Stretch-NICAM is larger than that in conventional regional models). We have added this to section 2.1 of the revised manuscript.

[C2-7] The scenario experiment is badly described and therefore difficult to follow. In its current form I find it superfluous to the manuscript as the results are very provisional and should be clearly declared as such. If the recommended improvements to the first part of the manuscript were made this would make a perfectly reasonable paper on its own without needing the future scenario experiment. It reduces the impact of this paper. The model configuration and method used to calculate the mortality rate is poorly referenced and insufficiently described. For example, I assume MIROC-AOGCM simulations are used to nudge Stretch-NICAM-SPRINTARS in August 2030 but this is not at all clear from the model description. What is "x" used in the calculation of D(x)? From the text I deduced that it doesn't refer to a NICAM grid point as the authors refer to a "NICAM grid" and "grid x" separately. Where were the population distributions taken from? I would recommend a total rewrite of Section 2.4 before publication is considered.

[A2-7] As you suggested, we have removed this part from the revised manuscript.

[C2-8] Large sections of the manuscript are poorly written and lack clarity making it difficult to follow the experimental design and subsequent evaluation. Given the focus on air quality a more detailed description of the aerosol scheme, in particular the sulphur chemistry is required in Section 2.2. I would recommend splitting Section 2.3 into 2 separate sections 1) Design of Experiment and 2) Observations.

[A2-8] Thank you very much for your comments. We have added the detail description of our aerosol model and the sulfur chemistry to section 2.2 of the revised manuscript. Could you directly check section 2.2 in the revised manuscript? Also, as you suggested, we have built "Design of the experiments" as section 2.3 and "Observations" as section 2.4 in the revised manuscript.

[C2-9] Furthermore the quality of the figures is very poor making it extremely difficult to follow the description in the manuscript.

[A2-9] Apologies for using the rainbow color without clear borderlines in figures. According to your suggestions, we have re-plotted all of figures.

Some specifics:

[C2-10] Section 2.2. L17: Are the authors assuming that all sulphate is in the form of ammonium sulphate?

[A2-10] Yes. We did not explicitly treat an internal mixture of sulfate and other species. Because this model cannot directly predict ammonium compounds, it is assumed that all sulfate is the form of ammonium sulfate. We have added this point to section 2.2 of the revised manuscript.

[C2-11] Section 2.2. L20: "The nitrate concentrations...can be disregarded" This is a confusing statement. Do the authors mean nitrate emissions are low enough in summertime in Japan to be disregarded in this study (in which case suitable references should be provided) or that nitrate is not represented in these simulations? Please rephrase for clarity.

[A2-11] Thank you very much for your comments. The nitrate mass concentrations in summer in the Kanto area are lower than the sulfate mass concentrations (Morino et al., 2010a). Therefore, in this study, to validate basic performance of NICAM-g6str as a general aerosol-transport model, we have focused on only sulfate as the representative secondary aerosol. We have modified this part in the revised manuscript as follows; "The nitrate in this study is disregarded, primarily because the main objective in this study is modeling of sulfate as a representative secondary aerosols and secondly because the nitrate mass concentrations are lower than the sulfate ones with the target of August 2007 in Japan (Morino et al., 2010c)".

[C2-12] Section 2.3: How long of a spin-up was allowed in the Stretch-NICAM-SPRINTARS simulations?

[A2-12] Although in the original manuscript, the spin-up time was just one week, we recalculated Stretch-NICAM-SPRINTARS with the spin-up time of one and a half months. As a result, the results in the revised manuscript were slightly changed from those in the original manuscript. Several sensitivity tests for the spinup time indicated that it is enough to set the spinup time to one and half month. We have added this to section 2.3 of the revised manuscript.

[C2-13] Section 2.4: "Therefore we combined Stretch-NICAM-SPRINTARS with MIROC_AOGCM by nudging..2026-2035". These sentences are badly constructed and very unclear. Please rephrase.

[A2-13] Thank you very much for your comments, but we have removed it.

[C2-14] Section 3.1: The description of Figure 8 does not reflect my interpretation of the same figure, where there are large discrepancies between model and observations. It is clear from Figures 8 and 9 that the model overpredicts the precipitation in the target Kanto region.

[A2-14] Thank you very much for your comments. As you mentioned, the model overpredicts the precipitation in the target Kanto region. We have inserted this point to the revised manuscript. We have modified the paragraph of the precipitation in the revised manuscript, as mentioned in the answer to your comment [A2-3].

[C2-15] Section 3.2.1, Last sentence: There is no evidence in the manuscript to support this statement that the simulations of trans-boundary pollution is well simulated. Remove or provide evidence.

[A2-15] Thank you very much for your comments. We have removed the sentence from the revised manuscript.

[C2-16] Figure 17 shows a clear underestimation in the extinction coefficient below 1km however they are within observational uncertainty. This should be stated as well as an explanation for the large uncertainty in the observations should be given.

[A2-16] Thank you very much for your suggestion. Surely, the extinction values observed by LIDAR include large variabilities, primarily because they are retrieved from the surface to the cloud base, which highly varies hour-by-hour and is basically difficult to detect with the high accuracy, and secondly because they depend not only on the PM2.5 mass concentrations but also on the ambient RH and the water amount attached to aerosols. We have added these comments to the revised manuscript.

[C2-17] Section 4.2: The role of nitrate in future emission scenarios is expected to increase and potentially outweigh SO2 emissions in terms of contribution (see for example Bellouin et al. JGR 2011 or Bauer et al. ACP 2007). Increasing emissions in Asia will therefore impact trans-boundary pollution in Japan and impact results found here. The limitations of this scenario study needs to emphasized.

[A2-17] Thank you very much for the comments. Although we have removed this part from the revised manuscript, we have added the limitation of our model caused by ignorance of nitrate under the future scenario experiment to summary of the revised manuscript.

[C2-18] Figure 19: I find it interesting that the MIROC-AOGCM shows higher regional variability in sulphate concentrations than NICAM given its coarser resolution. Do the authors have an explanation for this?

[A2-18] Thanks for your comments, but we have removed this part from the revised manuscript. The strong peak in the coarse resolution causes the higher variability in the sulfate concentrations.

[C2-19] Thank you for the opportunity to review this paper.

[A2-19] Thank you very much for giving variable comments and leading us to improve our manuscript.

1	Application of a global nonhydrostatic model with a
2	stretched-grid system to regional aerosol simulations
3	around Japan
4	

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26 Abstract

27An aerosol-coupled global nonhydrostatic model with a stretched-grid system has been 28developed. Circulations over the global and target domains are simulated with a single 29model, which includes fine meshes covering the target region to calculate meso-scale 30circulations. The stretched global model involves lower computational costs to simulate 31atmospheric aerosols with fine horizontal resolutions compared with a global uniform 32nonhydrostatic model, whereas it may require higher computational costs compared 33 with the general regional models, because the stretched-grid system calculates inside 34and outside the target domain. As opposed to general regional models, the stretched-grid system does require neither a nesting technique nor lateral boundary 3536conditions. In this study, we developed a new-type regional model for the simulation of 37aerosols over Japan, especially in the Kanto areas surrounding Tokyo, with a maximum 38horizontal resolution of approximately 10 km. This model usually reproduces temporal 39 variations and their averages of the observed weather around Japan. This model 40generally reproduces monthly mean distributions of the observed sulfate and SO2 over 41 East Asia, with the high correlations of more than 0.5, but the underestimation of the 42simulated concentrations by 40%. The underestimation is mainly caused by the 43underestimation in China and possibly by the uncertainty of the simulated precipitation around Japan. In the Kanto area, this model succeeds in simulating the wind patterns 4445and the diurnal transitions around the center of the Kanto area, although it is inadequate 46to simulate the wind patterns and the diurnal transitions at some sites located at the edge 47of the Kanto area and surrounded on three sides by mountains, e.g., Maebashi, mainly

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削除: We determined that this model was capable of
simulating meteorological fields and anthropogenic
primary particles, e.g., elemental carbon, and
secondary particles, such as sulfate, with comparable

with other regional models

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58	due to the insufficient horizontal resolution. This model also generally reproduces both
59	diurnal and weekly variations of the observed and/or a regional aerosol-transport model,
60	WRF-CMAQ, simulated EC, sulfate, and SO ₂ concentrations in the Kanto area,
61	especially with their high correlation (R>0.5) at Komae/Tokyo. Although the aerosol
62	module used in this study is relatively simplified compared to the general regional
63	aerosol models, this study reveals that our proposed model with the stretched-grid
64	system can be applicable for the regional aerosol simulation,

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削除: By combining the meteorological fields obtained from an atmosphere-ocean coupled model, we also applied the new model to a climate scenario experiment of PM2.5 (aerosol particles with diameters less than 2.5 μ m) over Japan with a high horizontal resolution to assess the public health impact at the prefecture scale.

72 **1** Introduction

Aerosols can greatly affect regional air quality and contribute to global climate change 7374(Forster et al., 2007). Recently, transboundary aerosol pollution, whereby regions 75beyond a given country's borders are affected by the aerosols generated in that country, 76has been of increasing concern (Ramanathan et al., 2008; Yu et al., 2012). The ongoing 77rapid economic growth in developing countries has the potential to exacerbate this issue 78(UNEP and WMO, 2011). Air pollution generated by aerosols is a critical public health 79issue due to the deleterious effects of these particles on human health (Dockery et al., 80 1993; Pope et al., 2009). Aerosols, which scatter and absorb solar radiation and act as 81 cloud condensation nuclei, can directly and indirectly change the Earth's radiation 82 budget. The majority of aerosols are emitted from localized areas, which are referred to 83 as hotspots, such as megacities and biomass-burning regions, and are spread throughout 84 the world via atmospheric transport (e.g., Ramanathan et al., 2008). Therefore, global 85 aerosol-transport models should consider the important regional-scale characteristics of 86 aerosol hotspots to reliably estimate their impacts on air quality and climate change.

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

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

98 For this purpose, we utilized the global Nonhydrostatic Icosahedral Atmospheric Model 99 (NICAM) developed by Tomita and Satoh (2004) and Satoh et al. (2008). NICAM has 100been employed for the global simulation of atmospheric processes with high-resolution 101grid spacing, whose size is comparable to the typical deep convective cloud scale. 102Miura et al. (2007) performed a one-week computation with a horizontal resolution of 1033.5 km using the Earth Simulator at the Japan Agency for Marine-Earth Science and 104Technology (JAMSTEC) to successfully simulate a Madden-Julian Oscillation (MJO) 105event. Suzuki et al. (2008) implemented an aerosol transport model named the Spectral 106Radiation-Transport Model for Aerosol Species (SPRINTARS; Takemura et al., 2005) 107in NICAM (we refer to this aerosol-coupled model as NICAM-SPRINTARS) and 108performed a one-week simulation with a horizontal resolution of 7 km using the Earth 109 Simulator. Although these global, highly resolved calculations are promising with 110regard to long-term climate simulations for decades, their requirement of vast computer 111 resources substantially limits their use in short-duration and/or case-specific simulations 112due to the current limitations of computational resources. To overcome this limitation, 113we adopt a compromise approach based on a new grid transformation named the

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

119 In this study, we focused on Japan, especially the Kanto region surrounding Tokyo 120 (Figure 1), because the Kanto region living more than 30 million people is one of the 121largest megacities in the world, In Japan, a monitoring system for the air pollution, e.g., 122PM2.5 (aerosol particles with diameters less than 2.5 μ m) and SO₂, has been operated 123by the Japanese government. Inorganic ions, mainly sulfate, have been measured over 124Japan and other Asian countries under EANET (Acid Deposition Monitoring Network 125in East Asia; http://www.eanet.asia/index.html). Measurements of carbonaceous 126aerosols were limited, with the exception of intensive measurements (Fine Aerosol 127Measurement and Modeling in Kanto Area, FAMIKA) in the Kanto region during 128summer 2007 (Hasegawa et al., 2008; Fushimi et al., 2011). For the model evaluation 129using these measurements, we simulated aerosol spatial distributions during August 1302007 using Stretch-NICAM-SPRINTARS with a horizontal resolution of approximately 13110 km over the Kanto region. Because the model framework of 132Stretch-NICAM-SPRINTARS is identical to that of globally uniformed grid simulation 133(we named it Global-NICAM-SPRINTARS), with the exception of the grid 134configuration, and involves lower computational costs than global simulations, the

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削除: and is located in eastern Japan (Figure 1)
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potentially vulnerable to air pollution. Within the
Kanto region

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	chemical compounds
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145	investigation of the model performance of Stretch-NICAM-SPRINTARS can be simply
146	and effectively extended to improve the original NICAM-SPRINTARS with globally
147	uniform high resolution for near-future simulations. To evaluate aerosol simulations
148	with the stretched-grid system, in this study we also conducted
149	Global-NICAM-SPRINTARS, but with relatively low resolution (approximately 100
150	km) due to the limited computational resources. Although the model inter-comparison
151	using different modules coupled to different dynamic cores cannot clarify the reasons of
152	the difference in the results among the models (e.g., Textor et al., 2006), the model
153	intra-comparison approach, with the exception of the grid system and the spatial
154	resolution, is very meaningful to investigate impacts of the stretched-grid system on the
155	aerosol simulations. In addition, Stretch-NICAM-SPRINTARS can be a new-type
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156	model that is also applicable for <u>a regional simulation</u> of aerosols, <u>because it focuses on</u>
156	model that is also applicable for <u>a regional simulation</u> of aerosols, <u>because it focuses on</u>
156 157	model that is also applicable for <u>a regional simulation</u> of aerosols <u>, because it focuses on</u> <u>a specific regional domain without</u> require a nesting technique <u>n</u> or boundary conditions,
156 157 158	model that is also applicable for <u>a regional simulation</u> of aerosols <u>, because it focuses on</u> <u>a specific regional domain without</u> require a nesting technique <u>n</u> or boundary conditions, unlike general regional models.
156 157 158 159	model that is also applicable for <u>a regional simulation</u> of aerosols <u>, because it focuses on</u> <u>a specific regional domain without</u> require a nesting technique <u>n</u> or boundary conditions, unlike general regional models. <u>For the model evaluation in the target Japan, we mainly focused on a representative</u>
156 157 158 159 160	model that is also applicable for <u>a regional simulation</u> of aerosols <u>because it focuses on</u> <u>a specific regional domain without</u> require a nesting technique <u>n</u> or boundary conditions, unlike general regional models. <u>For the model evaluation in the target Japan, we mainly focused on a representative</u> <u>primary aerosol, i.e., elemental carbon (EC), and a representative secondary aerosol, i.e.,</u>
156 157 158 159 160 161	model that is also applicable for <u>a regional simulation</u> of aerosols, <u>because it focuses on</u> <u>a specific regional domain without</u> require a nesting technique <u>n</u> or boundary conditions, unlike general regional models. For the model evaluation in the target Japan, we mainly focused on a representative primary aerosol, i.e., elemental carbon (EC), and a representative secondary aerosol, i.e., sulfate. EC is directly emitted from anthropogenic combustion processes, and is a good
156 157 158 159 160 161 162	model that is also applicable for <u>a regional simulation</u> of aerosols, <u>because it focuses on</u> <u>a specific regional domain without require a nesting technique <u>nor boundary conditions</u>, unlike general regional models. For the model evaluation in the target Japan, we mainly focused on a representative primary aerosol, i.e., elemental carbon (EC), and a representative secondary aerosol, i.e., sulfate. EC is directly emitted from anthropogenic combustion processes, and is a good indicator to monitor the transport pattern. The global and regional modelings for sulfate,</u>

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horizontal resolution than demonstrated in previous
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176	2010b). In addition, sulfate is the largest contributor to the total secondary inorganic
177	aerosols (e.g., Zhang et al., 2007), and the sulfate mass concentrations are larger than
178	that the nitrate ones in August 2007 over the Kanto area (Morino et al., 2010c).
179	Originally, these basic components (EC and sulfate) are suitable for the evaluation in
180	this study, primarily because the stretched-grid system was applied to the simulations of
181	atmospheric pollutants over the land in the mid-latitude bond for the first time and
182	secondly because the original SPRINTARS is more simplified compared to
183	conventional regional aerosol models.
184	This paper is organized as follows: the model framework of NICAM and SPRINTARS
185	and the experimental design are described in Section 2. We show two model results; (1)
186	Stretch-NICAM-SPRINTARS with glevel-6, in which "glevel" is the number of
187	divisions of an icosahedron used to construct the horizontal grid, (hereafter referred to
188	as the "NICAM-g6str" model) and (2) Global-NICAM-SPRINTARS with glevel-6
189	(hearafter referred to as the "NICAM-g6" model). In Section 3, the model results are
190	validated using in-situ measurements in terms of meteorological fields including
191	precipitation and aerosol species, especially EC, sulfate and SO ₂ . For the model
192	evaluation of chemical species, we also made use of results in a regional aerosol model,
193	the Community Multiscale Air Quality (CMAQ) driven by the Weather Research and
194	Forecasting (WRF) model named WRF-CMAQ, shown by Shimadera et al. (2013). We
195	also present the validation of total aerosol amounts, i.e., PM2.5, and aerosol optical

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削除: As a result, the simulations of transboundary air pollution, which is expected to increase in Asia (Takemura, 2012), are potentially superior to those obtained by general regional models. Given the heterogeneous distribution of populations in terms of the geography of megacities, Stretch-NICAM-SPRINTARS enables improved estimates of aerosol impacts on human health for future scenarios on a local scale, for example, within prefectures or municipalities of a country. Populations in megacities, particularly those in Asia, are highly susceptible to air pollution (UNEP and WMO, 2011). To predict the extent to which ambient particulates will affect the population in 2030, we performed a scenario experiment involving $\ensuremath{\mathsf{PM2.5}}$ (aerosol particles with diameters less than 2.5 μ m) around Japan by forcing Stretch-NICAM-SPRINTARS with meteorological fields obtained by an atmosphere-ocean coupled general circulation model (AOGCM), which is referred to as the Model for Interdisciplinary Research on Climate (MIROC) and was developed by the Atmosphere and Ocean Research Institute at the University of Tokyo (AORI/UT), the National Goto Daisuke 2014/10/29 14:33 削除: Stretch-Goto Daisuke 2014/10/29 14:33 削除:-Goto Daisuke 2014/11/21 12:57 削除: elemental carbon (Goto Daisuke 2014/11/21 12:57 **削除:**) Goto Daisuke 2014/10/27 17:01

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product, i.e., extinction for spherical aerosols, Finally, the conclusions are summarized in Section 4.

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261 2 Model description

262 **2.1** Nonhydrostatic Icosahedral Atmospheric Model (NICAM)

263NICAM, which employs an icosahedral grid-point method with a nonhydrostatic 264equation system (Tomita and Satoh, 2004; Satoh et al., 2008, 2014), is run with a 265maximum horizontal resolution of 3.5 km (Tomita et al. 2005; Miura et al., 2007) and 266can be applied to a transport model of aerosols and gases as a conventional atmospheric 267general circulation model (Suzuki et al., 2008; Niwa et al., 2011; Dai et al., 2014a, 2682014b; Goto, 2014). NICAM can also be employed for regional-scale simulations by 269adopting a stretched-grid system (Tomita, 2008a; Satoh et al., 2010). The stretched 270icosahedral grid was developed from a general grid transformation method, i.e., the 271Schmidt transformation method, for a horizontal grid system on a sphere. In the 272Schmidt transformation, the grid interval on a sphere lacks uniformity with a finer 273horizontal resolution close to the center of the target region. Tomita (2008a) showed 274that the Schmidt transformation minimizes potential errors involving the isotropy and 275homogeneity of the target region. The stretched-grid system can solve the main 276problems associated with commonly used regional models, which occur from artificial 277perturbations near boundary areas in cases where meteorological and aerosol fields are 278prescribed. In addition, the computational cost of the stretched-grid system is

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286	substantially lower than that of a global calculation under the same horizontal resolution
287	in the target region. For example, when the globally uniform grid with a maximum
288	horizontal resolution of 10 km is applied to the global simulation, the minimum
289	required theoretical computational cost is $64-256$ times higher than the cost of the
290	stretched-grid system in this study. Compared to conventional regional models, the
291	computational cost may increase because the stretched-grid system requires the
292	calculation outside the target domain. Furthermore, the model framework of the
293	stretched global model is identical to that of the uniformed global model without special
294	modifications, whereas the model framework of regional models is usually different
295	from that of global models. These advantages can facilitate additional developments for
296	global simulations by testing a new scheme with minimal computational cost.
297	Compared with general regional models, the stretched-grid system is more suitable for
298	the current study, which aimed to extend its use to the global uniform high-resolution
299	NICAM-SPRINTARS.

In this study, we adopt the stretched-grid system to focus on the Kanto region, including Tokyo, using glevel-6 resolution, and the stretched ratio of 100 (we call it <u>NICAM-g6str</u>), which is the ratio of the largest horizontal grid spacing located on the opposite side of the earth from Tokyo to the smallest horizontal grid spacing near Tokyo. As a result, a minimum horizontal resolution of 11 km around the center (140.00°E, 35.00°N) was used. NICAM implements comprehensive physical processes of radiation, boundary layer and cloud microphysics. The radiation transfer model is Goto Daisuke 2014/10/27 16:36

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311 implemented in NICAM with the k-distribution radiation scheme MSTRN, which 312incorporates scattering, absorption and emissivity by aerosol and cloud particles as well 313as absorption by gaseous compounds (Nakajima et al., 2000; Sekiguchi and Nakajima, 3142008). The vertical turbulent scheme comprises the level 2 scheme of turbulence closure 315by Mellor and Yamada (1974), Nakanishi and Niino (2004, 2009) and Noda et al. 316(2009). The cloud microphysics consist of the six-class single-moment bulk scheme 317(water vapor, cloud water, rain, cloud ice, snowflakes and graupel) (Tomita, 2008b). 318 Based on our experience in previous studies, we did not employ cumulus 319parameterization in this study (e.g., Tomita et al., 2005; Sato et al., 2009; Nasuno, 2013). 320 The topography used in this study is based on GTOPO30 (the horizontal resolution is 30 321arc seconds, that is approximately 1 km) courtesy of the U.S. Geological Survey. The 322vertical coordinates system adopts Lorenz grid and z* (terrain-following) coordinates 323with the 40 layers of z-levels and model top of 40 km height (Satoh et al., 2008), The 324timestep was set to 20 seconds.

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326 **2.2 SPRINTARS**

Based on the approach of Suzuki et al. (2008), the three-dimensional aerosol-transport model—Spectral Radiation-Transport Model for Aerosol Species (SPRINTARS; Takemura et al., 2000, 2002, 2005; Goto et al. 2011a,b,c)—was coupled to NICAM in this study. The SPRINTARS model calculates the mass mixing ratios of the primary tropospheric aerosols, i.e., carbonaceous aerosol (EC and OC, organic carbon), sulfate, soil dust, sea salt and the precursor gases of sulfate, namely, SO₂ and dimethylsulfide

337	(DMS). The aerosol module considers the following processes: emission, advection,
338	diffusion, sulfur chemistry, wet deposition and dry deposition, including gravitational
339	settling. For carbonaceous aerosols, the 50% mass of EC from fossil fuel sources is
340	composed of externally mixed particles, whereas other carbonaceous particles are
341	emitted and treated as internal mixtures of EC and OC (EC-OC internal mixture).
342	Biogenic secondary organic aerosols (SOAs) from terpenes are treated but are greatly
343	simplified by multiplying a conversion factor to the terpenes emission (Takemura,
344	<u>2012</u>). In addition, anthropogenic SOAs from toluene and xylene are disregarded in this
345	study. The bulk mass concentrations of EC, OC, and sulfate are calculated by
346	single-modal approach, which means that the SPRINTARS model does not explicitly
347	treat aerosol dynamic processes such as coagulation and condensation. The particle size
348	distribution of the dry particles are prescribed in a logarithmic normal size distribution
349	with dry mode radii of 18, 100, 80 and 69.5 nm, for pure EC, EC-OC internal mixture,
350	biogenic SOA and externally mixed sulfate, respectively (Goto et al., 2011a). The
351	hygroscopicities, densities and refractive indices for the aerosols are set to the same
352	values used by Takemura et al. (2002) and Goto et al. (2011a). The combinations of the
353	pre-calculated cross-sections of the extinction and simulated mixing ratios for each
354	aerosol species provide the simulated aerosol extinction coefficient for each timestep of
355	the model (Takemura et al., 2002). The sulfur chemistry in SPRINTARS considers only
356	three chemical reactions to form sulfate through gas-phase oxidation of SO ₂ by
357	hydroxyl radical (OH) and aqueous-phase oxidation by ozone and hydrogen peroxide.
358	The large part of SO ₂ are emitted from fossil fuel combustion, biomass burning, and

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366	volcano eruption, whereas some of SO_2 are formed from the oxidation of DMS, which
367	is emitted naturally from marine phytoplanktons. The numerical solution in the
368	oxidations adopts an approximation in a quasi first-order reaction using the same
369	integrated time resolution as that of the dynamic core. The pH value in the
370	aqueous-phase is fixed at 5.6, because the SPRINTARS model treats limited ions in the
371	aqueous-phase (e.g., Takemura et al., 2000). The oxidant distributions (OH, ozone and
372	hydrogen peroxide) were offline provided by a chemical transport model. The
373	atmospheric removal of aerosols in SPRINTARS includes wet (due to rainout and
374	washout) and dry (due to turbulence and gravity) deposition processes, whereas those of
375	SO ₂ only include rainout and dry deposition by turbulence. In the cloudy grid, the mass
376	fractions of sulfate out of the cloud droplets to the mass of sulfate in the grid were fixed
377	at 0.5, whereas the fractions for SO ₂ were determined by Henry's law (Takemura et al.,
378	2002a). As for pure EC, EC-OC internal mixture, and biogenic SOA, the mass fractions
379	were fixed at 0.1, 0.3, and 0.3, respectively. Because the SPRINTARS model does not
380	predict the mass mixing ratio of the chemical tracers inside the clouds, it assumes that
381	the tracers inside the clouds are evaporated from the clouds at one timestep. In this
382	study, the particle mass concentrations for diameters less than 2.5 μm (defined as
383	PM2.5) are calculated by summing EC, organic matter by multiplying OC by 1.6
384	(Turpin and Lim, 2001), sulfate and ammonium aerosols. Because this model cannot
385	directly predict ammonium compounds, it is assumed that all sulfate is the form of
386	ammonium sulfate, so that their concentration was estimated by multiplying the mass
387	concentration of sulfate by 0.27, which is the molar ratio of ammonium ion to

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392 ammonium sulfate. The nitrate in this study is disregarded, primarily because the main 393 objective in this study is modeling of sulfate as a representative secondary aerosols and 394 secondly because the nitrate mass concentrations are lower than the sulfate ones with

395the target of August 2007 in Japan (Morino et al., 2010c).

396

2.3 Design of the experiments 397

398 The target period comprises one month in August 2007, in which an intensive 399 measurement of aerosol chemical species was conducted under Project FAMIKA (Hasegawa et al., 2008; Fushimi et al., 2011). The six-hour meteorological fields (wind 400 401 and temperature) were nudged above a height of 2 km using NCEP-FNL reanalysis data 402(http://rda.ucar.edu/datasets/ds083.2/). The one-hour sea surface temperature was also 403 nudged using the NCEP-FNL data. The initial conditions were prescribed by the 404 NCEP-FNL data for the meteorological fields and the one and a half months spinup results of the Stretch-NICAM-SPRINTARS model for the aerosol fields, respectively. 405

406 The emission inventories of anthropogenic EC, OC and SO₂ in this experiment were 407 prepared by EAGrid2000 with a horizontal resolution of 1 km over Japan (Kannari et al., 408 2007), REAS version 2 with a horizontal resolution of 0.25° over Asia (Kurokawa et al., 409 2013) and the AeroCom inventory with a horizontal resolution of 1° over other areas of 410the world (Diehl et al., 2012). Because EAGrid2000 does not explicitly estimate EC and 411 OC inventories, we estimated the inventories to be consistent with those from previous 412studies (Morino et al., 2010a,b; Chatani et al., 2011) by modifying the PM2.5 inventory

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416 of EAGrid2000 using scaling factors of EC/PM2.5 and OC/PM2.5 based on sources.

417These inventories of anthropogenic EC and SO₂ in 2007 are described in Figure 2. The 418 emissions of SO₂ from volcanoes in Japan, such as Miyakejima and Sakura-jima, were 419 obtained from statistical reports (http://www.seisvol.kishou.go.jp/tokyo/volcano.html) 420by the Japan Meteorological Agency (JMA). In this study, the distributions of three 421hourly averaged monthly oxidants (OH, ozone and hydrogen peroxide) were derived 422from a global chemical transport model (CHASER) coupled to the Model for 423Interdisciplinary Research on Climate (MIROC), named MIROC-CHASER, with the 424spatial resolution of 2.8° by 2.8° (Sudo et al., 2002).

425To evaluate model performances in the stretched-grid system, we also simulated 426NICAM-SPRINTARS with the globally uniformed grid simulation in glevel-6 427resolution (the horizontal resolution is set to 110 km and we call it NICAM-g6). 428Global-NICAM-SPRINTARS with relatively low resolution has been applied to aerosol 429simulations and well compared with in-situ measurements and satellite remote sensing 430 (Dai et al., 2014a; Goto, 2014). In the NICAM-g6 simulation, the cloud physics apply 431both the prognostic Arakawa-Schubert-type cumulus convection scheme (Arakawa and 432Schubert, 1974) and the diagnostic large-scale clouds described by Le Treut and Li 433(1991). The large-scale cloud module is based on single moment bulk scheme for cloud 434mixing ratio. The precipitation rate is parameterized by Berry (1967). Except for the 435grid system and the horizontal resolution (which determines the module of the cloud physics), Global-NICAM-SPRINTARS was identical to Stretch-NICAM-SPRINTARS. 436

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Therefore, apart from general model inter-comparison projects including various
aerosol modules and dynamic cores, the comparison between NICAM-g6str and
NICAM-g6 led to clarify impacts of the horizontal resolution on the aerosol
distribution.

446

447 **2.4 Observation**

448In this study, we focused on the aerosol chemical component of EC as the primary 449particle and sulfate as the secondary particle. To evaluate the model results over the Kanto region, we used observations of the surface mass concentrations of EC and 450sulfate in four cities under Project FAMIKA: Maebashi/Gunma (139.10°E, 36.40°N), 451Kisai/Saitama (139.56°E, 36.09°N), Komae/Tokyo (139.58°E, 35.64°N) and 452453Tsukuba/Ibaraki (140.12°E, 36.05°N). The EC particles in PM2.5 were collected every 454six hours with quartz fiber filters and analyzed with the thermal/optical method 455according to the IMPROVE protocol (Chow et al., 2001). The sulfate particles in PM2.5 were also collected every six hours with Teflon filters and analyzed by ion 456457chromatography. In addition to the limited FAMIKA dataset, we utilized measurements taken by the EANET (Acid Deposition Monitoring Network in East Asia; 458459http://www.eanet.asia/index.html) and the 4th national survey report of acid rain over 460 fiscal 2007 in Japan vear (http://tenbou.nies.go.jp/science/institute/region/journal/JELA_3403041_2009.pdf) to 461462assess the monthly mean concentrations of sulfate and SO₂ at Japanese and Korean sites,

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467	Meteorological Administration Atmosphere Watch Network (CAWNET). To validate
468	the concentration of SO_2 for the Kanto region, we accessed monitoring stations operated
469	by Japanese and local governments.
470	In the validation of the meteorological fields simulated by <u>NICAM-g6str and</u>
471	NICAM-g6, we used meteorological fields (wind and temperature) reanalyzed by
472	NCEP-FNL over East Asia. In the Kanto region, we obtained measurements for the
473	meteorological parameters (temperature, relative humidity (RH) and wind) at or near
474	the <u>7</u> sites of Project FAMIKA and <u>additional</u> cities; Tsuchiura/Ibaraki (140.20°E,
475	36.07°N), which is the city nearest to Tsukuba; Yokohama/Kanagawa (139.64°E,
476	35.45°N); Chiba/Chiba (140.12°E, 35.62°N); Adachi/Tokyo (139.82°E, 35.77°N); and

We also obtained Chinese measurements by Zhang et al. [2012], as part of the Chinese

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477 Machida/Tokyo (139.43°E, 35.53°N), which is the city nearest to Komae, as shown in
478 Figure 1(b). For precipitation, we used a measurement taken by the Automated

479 Meteorological Data Acquisition System (AMeDAS) at <u>21 sites over Japan including</u>

480 the following 10 Kanto's sites: Yokohama; Chiba; Tsukuba; Tokyo, which is near

481 Adachi; Maebashi; Huchu, which is near Machida; Konosu, which is near Kisai; Abiko

482 (140.11°E, 35.60°N); Saitama (139.59°E, 35.88°N); and Nerima (139.59°E, 35.74°N)

(Figure 1). To evaluate the spatial patterns of the precipitation obtained by
 NICAM-g6str, and NICAM-g6, we used the quantities of the monthly mean

485 precipitation around Japan that were derived from the Global Satellite Mapping of

486 Precipitation (GSMaP; Okamoto et al., 2005; Kubota et al., 2007; Aonashi et al., 2009;

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493 Ushio et al., 2009) and the Meso Scale Model (MSM) developed by the JMA for rain
494 forecast (Saito et al., 2006). The results by MSM are generally higher accurate than
495 those in GSMaP, although the covering area in MSM is limited around Japan.

496 To evaluate the quantities of the total aerosol amounts, such as PM2.5, we compared the 497 simulated PM2.5 concentrations with the observations at the 18 sites including the 498 FAMIKA sites and other monitoring stations operated by the Japanese and local 499 governments (Figure 1). The PM2.5 concentrations were continuously observed using 500tapered element oscillating microbalance (TEOM) with Series 1400a Ambient 501Particulate Monitor, The instruments are controlled under the temperature of 50 °C, to 502minimize the influence of change in the ambient temperature and RH. However, it 503includes large uncertain due to the difficulty in completely eliminate the water content 504attached to aerosols and lacks of the calibration of the instrument in some of sites. 505Nevertheless, the observed PM2.5 concentrations with hourly time resolution were still 506useful to validate the model results.

In Tsukuba and Chiba, light detection and ranging (LIDAR) measurements operated by the National Institute for Environmental Studies (NIES) of Japan were also available (Sugimoto et al., 2003; Shimizu et al., 2004). The LIDAR unit measured vertical profiles of the backscattering intensity at 532 and 1064 nm and the depolarization ratio at 532 nm. The backscattering intensity was converted to the extinction coefficient, and the depolarization ratio distinguished the extinction between spherical and non-spherical particles. In this study, we only used vertical profiles of the extinction for spherical Goto Daisuke 2014/10/27 17:10 削除: forecast Grid Point Value (GPV) processed

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削除: of Kawasaki/Kanagawa, which is the city nearest to Yokohama; Machida/Tokyo; Koutou/Tokyo, which is site nearest to the site of Adachi/Tokyo; Osaka/Osaka (135.53°E, 34.68°N); Amagasaki/Hyogo (135.42°E, 34.72°N); and Nonodake/Miyagi (141.17°E, 38.55°N) Goto Daisuke 2014/10/31 14:37 削除:

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particles. A detailed algorithm was provided by Sugimoto et al. (2003) and Shimizu etal. (2004).

525

526 **3 Validation of Stretch-NICAM-SPRINTARS**

527 **3.1 Meteorological fields**

528So far, the stretched-grid system was mainly applied to the simulations of tropical 529cyclones or tropical convective clouds with small domain over oceans for the short-term 530period (less than several days) (e.g., Satoh et al., 2010; Arakane et al., 2013). In this 531study, we focused on the air pollution around Japan (for the longer period). Therefore, 532we first focused on the general circulation of the basic meteorological fields over the 533large domain, which can affect the air pollution over Japan. Figure 3 shows temperature 534and winds near the surface and the model height of approximately 5 km for the model 535bottom of MSL over Asia region (100°E-170°E, 10°N-50°N). In August, North Pacific 536High (or Ogasawasa High) mainly brings clear weather around Japan. A frequency of 537the precipitation is usually limited, but a total amount of the monthly mean precipitation 538is not small, because of typhoons and shower rain. In the focusing region, the general 539meteorological fields simulated by NICAM-g6str and NICAM-g6 are comparable to 540those obtained by NCEP-FNL. The absolute biases in the temperature between 541NICAM-g6str and NCEP-FNL or between NICAM-g6 and NCEP-FNL are within 5421.5 °C. At the model height of 5 km, the NICAM-g6str-simulated temperature tends to be larger than NICAM-g6-simulated one by at most 3 °C, probably because the spatial 543

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546	resolution in NICAM-g6str is finer than that in NCEP-FNL. These positive biases
547	between NICAM-g6str and NCEP-FNL can be seen around Japan. As for wind, western
548	winds over the northeastern part of Japan in both NICAM-g6str and NICAM-g6 are
549	stronger compared to those in NCEP-FNL. With the exception of this bias, the
550	performances of both NICAM-g6str and NICAM-g6 are good. Therefore, it is
551	concluded that the general circulations obtained by the stretched as well as the
552	uniformed grid systems are well reproduced under the nudging technique in this study.
553	To evaluate the model performances of the six-hourly instant concentrations of aerosol
554	chemical species and SO ₂ over the main target region, i.e., Kanto area, we used the
555	six-hourly instant observations of temperature, RH, wind and precipitation at each
556	station over the Kanto area shown in Figure 1, The results and summary are shown in
557	Figures 4 to 7 and Table 1. The NICAM-g6 results, especially in terms of diurnal
558	variations, tend to be far from the observations compared to the NICAM-g6str results,
559	because NICAM-g6, with the horizontal resolution of approximately 100 km, does not
560	fully resolve the topology over the Kanto area. Figure 4 illustrates the temporal
561	variations of temperature at a height of 2 m. The temporal variations in the
562	NICAM-g6-simulated temperature are generally comparable to those in the observed
563	temperatures with root-mean-square-error (RMSE) values of less than 3°C, with the
564	exception of the results obtained for Maebashi and Machida. At these two sites, the
565	mean values of the NICAM-g6str-simulated temperatures are lower than those of the
566	observed temperatures by a maximum of 3.6° C. The correlation coefficients (R)

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572	between <u>NICAM-g6str</u> and <u>the</u> observation range from 0.7–0.9, <u>whereas the R between</u>	 Goto Daisuke 2014/11/18 18:49	
573	NICAM-g6 and the observation range from 0.7-0.8, as shown in Table 1. Figure 5	削除: theICAM-g6str simulationsnd the	3
574	shows the temporal variations in RH at a height of 2 m. The temporal variations in the		
575	NICAM-g6str-simulated RH are similar to the observations, with the RMSEs in the		
576	range of 10-15%. In contrast, the NICAM-g6-simulated RH is overestimated compared		
577	to the observations, with the RMSEs in the range of 16-26%. The R values of RH		
578	between the simulation (both NICAM-g6str and NICAM-g6) and observations are		
579	approximately 0.6–0.8 (Table 1).		
580	The temporal variations in the wind direction and speed simulated by <u>NICAM-g6str</u> are	Goto Daisuke 2014/11/14 16:39	
581	compared with the observations in Figures 6 and 7. Near the southern part of the Kanto	削除: Stretch-NICAM-SPRINTARSICAM- <u>e6</u>	tr ID
582	area (Yokohama, Tsuchiura, Adachi and Machida), with the exception of Chiba, the		
583	NICAM-g6str-simulated wind direction, is generally comparable to the observations,		
584	with a slight overestimation of the both NICAM-g6str and NICAM-g6 simulated wind		
585	speed compared with the observations. At these four sites, the R and RMSE values in		
586	NICAM-g6str range from approximately 0.5–0,7 and approximately 1,7–2,3 m/s,		
587	respectively. In Chiba located near the ocean, the R value of wind speed between		
588	NICAM-g6str, and the observation, is 0,41, whereas the NICAM-g6str-simulated wind		
589	directions generally agree with the observations. Conversely, at Maebashi and Kisai, the		
590	daily variations in the both NICAM-g6str and NICAM-g6 simulated wind directions		
591	differ significantly from those in the observations, in which the southern winds and		
592	northern winds frequently occur during the day and night, respectively, for example,		

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615	during August 5-12. At these two sites, the NICAM-g6-simulated wind direction and
616	speed is not closer to the observations compared to those obtained by NICAM-g6str.
617	The R value for wind speed between the <u>NICAM-g6str</u> and <u>the</u> observations at these
618	sites is estimated to be approximately 0.2. The observed southeasterly wind is long sea
619	breeze toward Maebashi Plateau surrounded on three sides by mountains around
620	Maebashi. The observed winds are caused by daytime meso-scale thermal lows
621	developed over the central Japan covering the Japanese Alps (Kuwagata and Sumioka,
622	1991). The Japanese Alps with the highest terrain in Japan can affect the local
623	meteorological fields even around 100-200 km away (Kitada et al., 1998). Therefore, it
624	suggests that the horizontal resolution in this study using NICAM-g6str (10 km over the
625	Kanto area) does not fully resolve the complex terrains of the Japanese Alps and the
626	Maebashi plateau. Therefore, it suggests that it is inadequate to simulate the wind
627	patterns and the diurnal transitions near high mountains, around the Kanto area, whereas
628	it is adequate to simulate them around the center of the Kanto area.
629	Figures 8-10 show, comparisons of NICAM-g6str and NICAM-g6 simulated
630	precipitation with the observations. Figure 8 compares the simulated precipitation with
631	the MSM and GSMaP derived results. During the early August 2007, mainly due to
632	passing of a typhoon over the western Japan, Okinawa, and Korea, the August mean
633	precipitation in the western Japan is larger than that in the eastern Japan, especially the
634	Kanto area. The monthly mean precipitation is estimated to be more than 200
635	mm/month over the western Japan, whereas that is estimated to be less than 50

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Maebashi and Kisai, which are surrounded by or are
located relatively close to high mountains, indicate
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study using Stretch-NICAM-SPRINTARS could not
completely resolve the topography. As a result,
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precipitation per day at each site. During August
2007 in the Kanto region, the observed precipitation
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672	mm/month over the eastern Japan. The horizontal patterns of the precipitation obtained
673	by NICAM-g6str in East China Sea, Sea of Japan near the Japan coast, and Korea are
674	closer to those derived from MSM and GSMaP than those obtained by NICAM-g6. In
675	the Kanto area, however, the NICAM-g6str-simulated precipitation with the range of
676	50-200 mm/month is overestimated compared to the MSM and GSMaP results. The
677	NICAM-g6-simulated precipitation over the Kanto area with the range of 100-200
678	mm/month is also much overestimated. In Figure 9 showing the temporal variations in
679	the amount of precipitation per day at 21 Japanese sites, the observed precipitation is
680	extremely limited during August 7-19 in the Kanto area. In other regions, the magnitude
681	of the precipitation is strong, although the precipitation is sporadic. In terms of the
682	frequency of the precipitation, the NICAM-g6str performance is better than the
683	NICAM-g6 one. Figure 10 illustrates the predictive value of daily precipitation, defined
684	as the ratio of the number of days where the model correctly predicts the weather (less
685	than 1 mm/day or more than 1 mm/day) to the number of the whole days. In the
686	NICAM-g6str results, the predictive values at most of sites over the Kanto area and four
687	sites over the non-Kanto area such as Nagoya and Osaka are calculated to be more than
688	85%. The predictive values obtained by NICAM-g6-str are mostly higher than those
689	obtained by NICAM-g6. During the rainy days such as August 20, 22 and 23 over the
690	Kanto area, both NICAM-g6str and NICAM-g6 capture the precipitation, whereas
691	NICAM-g6str reproduces greater amounts of the precipitation and NICAM-g6
692	reproduces longer periods and larger areas compared to the observations. NICAM-g6str
693	does not always capture a sudden shower, as general meteorological models have

694	difficulties in predicting this type of precipitation system (e.g., Kawabata et al., 2011).	
695	To increase the accuracy of such precipitation, more sophisticated cloud-microphysics	
696	model, e.g., NICAM-NDW6 model proposed by Seiki and Nakajima (2014) based on	
697	the double-moment bulk scheme with six water categories, may be required. In the	G
698	western Japan, during the rainy days, e.g., August 22-23, both NICAM-g6str and	削 ot
699	NICAM-g6 usually capture large-scaled precipitation (Figure 9). Overall, NICAM-g6str	w
700	usually reproduces the observed weather in the target regions and periods with a large	K ₂ ge
701	uncertainty, whereas NICAM-g6 does not capture general feature such as the sporadic	he
702	precipitation.	of Ol
703		N
704	3.2 Aerosol fields	G 削
705	3.2.1 Evaluation of chemical species,	G
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706	Figures <u>11, 12, and 13</u> illustrates the temporal variations in the surface EC, sulfate, and	G 削
707	SO ₂ concentrations at the four stations (Maebashi, Kisai, Komae and Tsukuba) in the	G 削
708	Kanto area using the simulations and the measurements. The simulations include	G
709	NICAM-g6str, NICAM-g6, and the Community Multiscale Air Quality (CMAQ) driven	削 G
710	by the Weather Research and Forecasting (WRF) model named WRF-CMAQ shown by	削
711	their Figures 5 and 6 of Shimadera et al. (2013). Shimadera et al. (2013) calculated the	G 削
712	WRF-CMAQ with a horizontal resolution of 5 km and an emission inventory that is	G 削
713	similar to that in the present study. Table 2 summarizes the statistical parameters for the	G
714	<u>concentrations of EC, sulfate, and SO₂.</u> The temporal variation and the average of EC	削 G

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除: The overestimation of the precipitation tained by Stretch-NICAM-SPRINTARS compared ith the observations is also seen in the Sea of Japan, yusyu, and the main island of Japan. All results nerally show similar patterns of the occurrence of avy precipitation in the East China Sea and the Sea Japan near the Japan coast, especially near kinawa, the southern part of South Korea and orth Korea. Therefore oto Daisuke 2014/10/29 15:13 除: Stretch-NICAM-SPRINTARS oto Daisuke 2014/11/18 18:56 除: can generally oto Daisuke 2014/11/18 18:56 除: simulate oto Daisuke 2014/10/29 15:13 除: meteorological fields oto Daisuke 2014/11/18 18:56 除: present oto Daisuke 2014/11/8 14:57 除: using measurements oto Daisuke 2014/11/8 14:57 除: . oto Daisuke 2014/11/8 14:57 **除:** 10 oto Daisuke 2014/11/8 14:57 除: mass oto Daisuke 2014/11/18 18:56 削除: obtained by Project FAMIKA

734	simulated by NICAM-g6str are better agreement with the observations obtained for
735	Komae than those simulated by NICAM-g6 (Figure 11(c)). However, the averages of
736	both NICAM-g6str and NICAM-g6 simulated EC concentrations at the other sites are
737	much underestimated compared to the observations (Table 2). For Tsukuba, shown in
738	Figure <u>11(d)</u> , <u>both the NICAM-g6str and NICAM-g6</u> simulated EC concentrations tend
739	to be underestimated compared with the observed concentrations, especially during the
740	daytime, even though the temporal variation of EC obtained by NICAM-g6str is closer
741	to the observed one compared to those obtained by NICAM-g6. At Maebashi and Kisai,
742	the temporal variation, and the averages of EC obtained by NICAM-g6 are also
743	underestimated compared with the observations by a factor of three to five.
744	NICAM-g6str tends to have daily maximums of EC concentrations during the morning
745	time, whereas NICAM-g6 tends to have daily maximums during the nighttime. The
746	temporal variations of NICAM-g6str-simulated EC concentrations are generally
747	comparable to those by WRF-CMAQ shown in Figure 11 and their Figure 3 of Chatani
748	et al. (2014), with the exception of the results at Maebashi and Kisai where the EC
749	concentrations obtained by NICAM-g6str are smaller than those obtained by
750	WRF-CMAQ. At these sties, the difference in the EC concentrations between
751	NICAM-g6str_and_WRF-CMAQ is probably caused by the difference in the horizontal
752	resolution, which is most likely critical for properly simulating the air pollution
753	delivered by the meteorological wind fields from the center of the Kanto region (Kusaka
754	and Hayami, 2006). Table 2 also shows that the R obtained by NICAM-g6str at all sites
755	are high or moderate, with the exception of Maebashi, whereas those obtained by

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comparable with the observations. Conversely
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772	NICAM-g6 and CMAQ are low. At most sites, the EC concentrations obtained by
773	WRF-CMAQ shown in Figure 11, and WRF-CMAQ illustrated by Morino et al.
774	(2010a,b) and Chatani et al. (2014), NICAM-g6str, and NICAM-g6 are also
775	underestimated compared to the observations with the larger values of RSME. The
776	underestimation of EC concentrations is investigated by sensitivity tests of EC emission
777	inventory in section 3.2.2.
778	At the same four sites, simulated sulfur components (sulfate and SO ₂) are compared
779	with the observations in Figures 12 and 13 . The observed SO ₂ represents the ensemble
780	results of monitoring stations operated by Japanese and local governments around each
781	FAMIKA site. The <u>mean</u> differences in the sulfate mass concentrations between
782	NICAM-g6str, and the observations are within approximately 10% at Maebashi and
783	Tsukuba, approximately -30% at Komae, and approximately +40% at Kisai. At all sites,
784	the temporal variations of the <u>NICAM-g6str-simulated</u> sulfate <u>concentrations</u> are
785	generally comparable to those <u>obtained by</u> the observations and WRF-CMAQ shown in
786	Figure 12 (i.e., their Figure 6 of Shimadera et al., 2013) and illustrated in their Figure 3
787	of Morino et al. (2010a), whereas the differences in the sulfate concentrations between
788	NICAM-g6str and the observations, are somewhat greater on August 7 and 8 at
789	Maebashi where the performance of NICAM-g6str is relatively poor, mainly due to the
790	inadequate horizontal resolution to reproduce the observed meteorological fields, as
791	shown in section 3.1, The use of the prescribed distributions of three hourly averaged
792	monthly oxidants may partly cause the discrepancy of the hourly variations of the

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26

809	sulfate between NICAM-g6str and the observations. The R obtained by all the models
810	(NICAM-g6str, NICAM-g6, and WRF-CMAQ) is acceptable at most sites, with the
811	exception of NICAM-g6str at Maebashi and WRF-CMAQ at Kisai. The RMSEs
812	obtained by all the models are smaller at Komae and Tsukuba than those at Maebashi
813	and Kisai. The six-hourly variations of the sulfate obtained by WRF-CMAQ are
814	sometimes missed by NICAM-g6str, partly due to the use of the prescribed oxidants.
815	Even though NICAM-g6 reproduces the weekly cycle of the observed sulfate, it has
816	difficulties in simulating the diurnal cycle of the observed and NICAM-g6str-simulated
817	sulfate, as shown in the results of EC by Figure 11. The averages of the sulfate
818	concentrations obtained by NICAM-g6 tend to be smaller than those by NICAM-g6str
819	and the observations. The possible impacts of the prescribed oxidant on the sulfate
820	concentrations are investigated in section 3.2.2.
821	In Figure 13, NICAM-g6str and NICAM-g6 simulated SO ₂ concentrations are
822	compared by the observations. In the previous studies, the comparison in SO_2
823	concentrations between the simulation and observation was very limited, with the
824	exception of their Figure 4 of Morino et al. (2010b), which showed large differences in
825	the SO ₂ concentrations between WRF-CMAQ and the observations by more than a
826	factor of two. The R between NICAM-g6str and the observations are low, with the
827	exception of Komae (R=0.62), but are approximately within the range obtained by
828	WRF-CMAQ in Morino et al. (2010b). The differences in the mean SO ₂ concentrations
829	between <u>NICAM-g6str and the observations and between NICAM-g6 and the</u>

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832	observations are within approximately 20% at all sites, with the exception of
833	NICAM-g6str at Maebashi and NICAM-g6str at Tsukuba (Table 2). The temporal
834	variations in the simulated SO_2 concur with those in the observations. <u>The observations</u>
835	sometimes show high SO ₂ concentrations at all sites, e.g., up to 20 ppbv at Komae, in
836	the afternoon on August 12 and 14. On August 12, NICAM-g6str normally reproduced
837	the peaks of the observed SO2 but with the blunter and slightly shifted peaks. On
838	August 14, both NICAM-g6str and NICAM-g6 did not reproduce the sharp peaks of the
839	observed SO ₂ , especially at Komae and Tsukuba. It may imply that special
840	meteorological fields cause the observed peaks on August 12, whereas local SO_2
841	emission is stronger on August 14. The latter issue is improved by processing
842	time-highly-resolved emission inventories of SO ₂ , which can be estimated through a
843	top-down approach using a data assimilation (Schutgens et al., 2012; Xu et al., 2013).
844	To assess the performance of both NICAM-gs6tr and NICAM-g6, in simulating the
845	distributions of the air pollutants over Japan, we compared the August averages of the
846	simulated <u>EC</u> , sulfate and SO ₂ <u>concentrations</u> with the available measurements (Figures
847	,14 and 15). Although the EC observatories are limited, both the NICAM-g6str and
848	NICAM-g6 simulated EC concentrations are much underestimated compared to the
849	observations, with the relative bias (Br) , defined as a ratio of the simulated value to the
850	observed one, to be 0.15 (NICAM-g6str) and 0.16 (NICAM-g6). In China, the
851	NICAM-g6str-simulated EC concentrations are comparable to the
852	NICAM-g6-simulated ones with the R values of 0.71 (NICAM-g6str) and 0.68

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860	(NICAM-g6), whereas at the Japanese urban areas such as Nagoya (136.97°E, 35.17°N)
861	and Osaka (135.54°E, 34.68°N), the NICAM-g6str-simulated EC concentrations are
862	larger than NICAM-g6-simulated ones.
863	The NICAM-g6str-simulated sulfate concentrations are larger and more comparable to
864	the observations over China compared to NICAM-g6-simulated ones. In Japan, the hot
865	spots with greater concentrations of more than 5 μ g/m ³ are found only in the
866	NICAM-g6str results. The Br values are estimated to be 0.59 (NICAM-g6str) and 0.53
867	(NICAM-g6), whereas the R values are estimated to be 0.78 (NICAM-g6str) and 0.88
868	(NICAM-g6), respectively. The results indicate that the sulfate concentrations obtained
869	by both NICAM-g6str and NICAM-g6 tend to be underestimated by approximately
870	40-50% compared with the observed sulfate concentrations. The underestimation is
871	mainly caused by the underestimation in China and possibly by the uncertainty of the
872	simulated precipitation around Japan. At Hedo located at Okinawa islands, for example,
873	the underestimation of both NICAM-g6str and NICAM-g6 simulated sulfate
874	concentrations is caused by a possible underestimation of transboundary sulfate from
875	the continent, which is attributed to a large uncertainty of the precipitation fields
876	modulated by typhoon in the early August. However, the correlations between the
877	simulations and observations are adequately acceptable, It suggests that the use of the
878	prescribed oxidants for sulfate formation is not crucial for predicting monthly averaged
879	sulfate mass concentrations at least if the diurnal and seasonal variations of the
880	prescribed oxidants are considered. The simulated and observed SO ₂ concentrations also

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890	correlate, with the R value of 0.63 (NICAM-g6str) and 0.48 (NICAM-g6), The Br
891	values are calculated to be 0.48 (NICAM-g6str) and 0.67 (NICAM-g6). Figure 15
892	shows that the SO_2 , which is a primary product, is localized near the source areas,
893	whereas sulfate, which is as a secondary product, is distributed from the source to the
894	outflow areas. Although EC is also a primary product, the horizontal distributions of
895	NICAM-g6str-simulated EC are smaller than those of NICAM-g6str-simulated SO2.
896	possibly because SO ₂ near the surface is more scavenged through the dry deposition
897	process compared to EC,

899 **3.2.2 Uncertainty in the simulation**

900 Sensitivity tests were conducted to examine potential uncertainties derived from 901 prescribed datasets related to EC and sulfate for the NICAM-g6str simulations. For the 902 EC sensitivity tests, the emission quantities were set to half and twice of those used in 903 the standard run in this study. The results for the FAMIKA sites are shown in Figure 904 16(a) in which the bars show the simulated EC concentrations for both sensitivity tests. 905For the majority of the sites, with the exception of Komae, the results obtained by the 906 sensitivity experiments of twice strength remain underestimated compared with the 907 measurements. The large underestimation of the EC mass concentrations at Maebashi 908 and Kisai was also shown by WRF-CMAQ of Shimadera et al. (2013) as well as the 909 previous studies of WRF-CMAQ in Morino et al. (2010a,b) and Chatani et al. (2014), 910 However, Fushimi et al. (2011) and Chatani et al. (2014) suggested that the difference

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	2007. The
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	Community Multiscale Air Quality (CMAQ) driven
	by the Weather Research and Forecasting (WRF)
	model named WRF-CMAQ with a horizontal
	resolution of 5 km. WRF-CMAQ employs an
	emission inventory that is similar to that in the
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/	concentrations at Maebashi between the present
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954	in the EC concentrations between WRF-CMAQ and the measurements is largely	
955	attributed to an underestimation of the EC emission inventory, especially open biomass	
956	burning from domestic sources. The local EC emission can be estimated by a	
957	combination of the data assimilation and intensive measurements (Schutgens et al.,	
958	2012; Wang et al., 2012; Yumimoto and Takemura, 2013),	
959	Sensitivity experiments of the SO_2 emissions and the prescribed <u>OH</u> radical used in	/
960	sulfur chemistry were executed under half and twice the amounts used in the standard	
961	experiment. The results for the FAMIKA sites are shown in Figure 16(b) in which the	/
962	bars show the simulated sulfate concentrations for both sensitivity tests under the	
963	different experiments. Compared with the SO2 emissions used in the standard	
964	experiment, the doubled amount of SO_2 emissions can overcome the slight	
964 965	experiment, the doubled amount of SO_2 emissions can overcome the slight underestimation of the simulated sulfate compared with the observations. Therefore, the	
965	underestimation of the simulated sulfate compared with the observations. Therefore, the	
965 966	underestimation of the simulated sulfate compared with the observations. Therefore, the emission inventories of SO ₂ should be improved for the better simulation of the sulfate.	
965 966 967	underestimation of the simulated sulfate compared with the observations. Therefore, the emission inventories of SO_2 should be improved for the better simulation of the sulfate. In this sensitivity tests for oxidants, the SO_2 oxidation by OH radical strongly depends	
965 966 967 968	underestimation of the simulated sulfate compared with the observations. Therefore, the emission inventories of SO_2 should be improved for the better simulation of the sulfate. In this sensitivity tests for oxidants, the SO_2 oxidation by OH radical strongly depends on the OH concentrations as well as the cloud cover area, whereas the SO_2 oxidation by	
965 966 967 968 969	underestimation of the simulated sulfate compared with the observations. Therefore, the emission inventories of SO_2 should be improved for the better simulation of the sulfate. In this sensitivity tests for oxidants, the SO_2 oxidation by OH radical strongly depends on the OH concentrations as well as the cloud cover area, whereas the SO_2 oxidation by ozone and hydrogen peroxide mainly depends on their concentrations, the cloud cover	
965 966 967 968 969 970	underestimation of the simulated sulfate compared with the observations. Therefore, the emission inventories of SO_2 should be improved for the better simulation of the sulfate. In this sensitivity tests for oxidants, the SO_2 oxidation by OH radical strongly depends on the OH concentrations as well as the cloud cover area, whereas the SO_2 oxidation by ozone and hydrogen peroxide mainly depends on their concentrations, the cloud cover area, and the cloud water content. The cloud distributions are modulated by some	
965 966 967 968 969 970 971	underestimation of the simulated sulfate compared with the observations. Therefore, the emission inventories of SO_2 should be improved for the better simulation of the sulfate. In this sensitivity tests for oxidants, the SO_2 oxidation by OH radical strongly depends on the OH concentrations as well as the cloud cover area, whereas the SO_2 oxidation by ozone and hydrogen peroxide mainly depends on their concentrations, the cloud cover area, and the cloud water content. The cloud distributions are modulated by some feedbacks of the sulfate formation through the aerosol direct and indirect effects. As a	

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削除: Therefore, the same factor may be applicable to the present results using Stretch-NICAM-SPRINTARS Goto Daisuke 2014/11/18 19:20 削除: hydroxyl Goto Daisuke 2014/11/14 17:16 移動 (挿入) [4] Goto Dais 削除: Figure 15(b) shows that the sensitivity of the hydroxyl radical concentrations to the simulated sulfate concentration is substantially smaller than that to the SO_2 emissions. We also determined that the sensitivities of the other oxidants to the simulated sulfate concentrations were minimal (not shown). Goto Daisuke 2014/11/14 17:16 上へ移動 [4]: We also determined that the sensitivities of the other oxidants to the simulated sulfate concentrations were minimal (not shown). Goto Daisuke 2014/11/14 17:26 削除: These results from the sensitivity experiments indicate that the offline prescribed oxidant used in this study is not as critical to the proper prediction of the sulfate concentrations over the Kanto region as the uncertainty in the quantity of SO2 emissions. Goto Daisuke 2014/11/18 19:21 削除: we conclude that the simulations of Stretch-NICAM-SPRINTARS are generally successful in simulating the air pollution over Japan and are adequate as a new regional model for simulations over the Kanto region

998	concentrations were small (not shown). These results also suggest that the use of the		
999	prescribed oxidants for sulfate formation is not crucial for predicting weekly-averaged		
1000	sulfate mass concentrations at least by taking into account for diurnal and seasonal		
1001	variations of the prescribed oxidants. At the same time, they also suggest that the		
1002	relationship between the oxidants and the sulfate concentrations through the feedbacks		
1003	is non-linear and complex, and thus the sensitivity of the oxidants to the simulated		G
1004	sulfate should be investigated.		削 St
1005			G
1006	3.2.3 PM2.5	V ,	削 G
1007	Figure $\frac{17}{10}$ shows the temporal variation in the surface PM2.5 mass concentration at the		削 G
1008	<u>18 Japanese sites including 10 sites in the Kanto area.</u> At most of the sites, both		削
1000	<u>10 Japanese</u> sites <u>melading registes in the</u> Kanto <u>area</u> . At <u>most of the</u> sites, <u>both</u>	$\overline{\left\langle \right\rangle}$	G
1009	NICAM-g6str and NICAM-g6 usually captures the weekly variation of the observed		削 G
1010	PM2.5, whereas only NICAM-g6str reproduces the diurnal variation of the observed		削
1011	PM2.5. Table 3 shows the PM2.5 concentrations in daily, daytime (from 9 am to 4 pm),		G 们
1012	and nighttime (from 9 pm to 4 am) averages and ratios of daytime to nighttime. The		G
1013	results show that the simulated PM2.5 concentrations are underestimated compared with		削 PN
1014	the observations by more than a factor of two and by up to four at Maebashi. In addition,		va
1015	the results show that the NICAM-g6str-simulated ratios (0.9-1.3) are larger than		G 削
1016	NICAM-g6-simulated ones (0.8-0.9), whereas the NICAM-g6str-simulated ones are		G 削
1017	smaller than the observed values (1.0-1.8). At Maebashi, where the ratio is higher than		G
1018	that at other sites, the issue of the poor model performance of the meteorological fields		削 G

oto Daisuke 2014/11/8 15:14 除: 4 Application of tretch-NICAM-SPRINTARS[10] oto Daisuke 2014/11/8 15:14 除: 1 oto Daisuke 2014/11/8 15:14 **除:** 16 oto Daisuke 2014/11/8 15:15 **除:** 11 oto Daisuke 2014/11/8 15:15 除: over the oto Daisuke 2014/11/8 15:15 除: region and in western and northern Japan oto Daisuke 2014/11/8 15:15 除: all oto Daisuke 2014/11/8 15:16 除: the temporal variations in the simulated M2.5 are generally similar to those in the observed lues; however, oto Daisuke 2014/11/8 15:16 除: or three at the majority of sites oto Daisuke 2014/11/8 15:16 除: approximately oto Daisuke 2014/11/8 15:16 除: a factor of oto Daisuke 2014/11/8 15:16 削除: In addition to

1035	can be a major reason of the large underestimation, as mentioned in section 3.1. At all
1036	sites, the possible underestimation of SOA, may be a critical issue, as shown in the fact
1037	that the clear diurnal variation of PM2.5 during August 4-9 and suggested by previous
1038	studies (Matsui et al., 2009; Morino et al., 2010c). Morino et al. (2010c) implied that
1039	over the Kanto area SOA from anthropogenic sources, which were disregarded in this
1040	study, are large portion of total carbonaceous aerosols, even though WRF-CMAQ does
1041	not correctly reproduce such carbonaceous aerosols. More sophisticated SOA module,
1042	e.g., volatility basis-set approach proposed by Donahue et al. (2006) based on the
1043	categorization of organic vapors with similar volatility, is required for to produce SOA
1044	with higher accuracy. Originally, the underestimation of PM2.5 is common among
1045	previous studies that employed regional aerosol-transport models (Morino et al., 2010b,
1046	Chatani et al., 2011), primarily because the concentrations of the observed PM2.5
1047	include undefined chemical species with mean fractions ranging from approximately
1048	30-50% in the total PM2.5 in the summer of Japan (datasets from the Tokyo
1049	Environment Agency and the Kawasaki Municipal Research Institute for Environmental
1050	Protection). Another possible reason is that the PM2.5 mass concentration includes
1051	water attached to aerosols, depending on the ambient RH conditions. Therefore, these
1052	undefined chemical compounds in this study may account for a large portion of the
1053	difference between the simulated and the observed values.
1054	To evaluate the vertical profiles of the PM2.5 mass concentrations, we used the LIDAR
1055	observation operated by the NIES-Japan network. Figure <u>18</u> shows the average results

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	下へ移動 [2]: previous studies that employed
	regional aerosol-transport models (Morino et al.,
	2010b, Chatani et al., 2011)
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	移動 (挿入) [2]
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1065	for the simulated and observed extinction coefficient of the spherical particles at
1066	Tsukuba and Chiba in August. At both sites, the vertical profiles and the magnitudes
1067	below 3 km height of the simulated extinction by both NICAM-g6str and NICAM-g6
1068	are comparable to the observed results, whereas the simulated extinction values tend to
1069	be smaller than the observed extinction values near the surface. These results near the
1070	surface are consistent with those obtained by the surface PM2.5 comparison shown in
1071	Figure 17. In contrast, the extinction values observed by LIDAR include large
1072	variabilities, primarily because they are retrieved from the surface to the cloud base,
1073	which highly varies hour-by-hour and is basically difficult to detect with the high
1074	accuracy, and secondly because they depend not only on the PM2.5 mass concentrations
1075	but also on the ambient RH and the water amount attached to aerosols. At both sites, the
1076	differences in the extinction between NICAM-g6str and NICAM-g6 are small below 1
1077	km height, whereas those are relatively large above 1 km height. The differences are
1078	attributed to the differences in the primary particles, mainly carbonaceous aerosols,
1079	between NICAM-g6str and NICAM-g6 (not shown). It means that it is attributed to the
1080	difference in the vertical transport between different spatial resolutions. Therefore,
1081	impacts of the difference in the spatial resolution on the distributions of both aerosols
1082	and their precursors, should be addressed in the future work,
1083	
1084	A Summary

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1	Goto Daisuke 2014/11/8 15:19
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	Stretch-NICAM-SPRINTARS are used in an
	estimation of health impacts due to PM2.5, the bias
١	Goto Daisuke 2014/11/8 15:19
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1	Goto Daisuke 2014/11/8 15:20
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	impact .

1096	An aerosol-coupled global nonhydrostatic model, which is based on the aerosol module
1097	of Spectral Radiation-Transport Model for Aerosol Species (SPRINTARS) and the
1098	global cloud-resolving model of Nonhydrostatic Icosahedral Atmospheric Model
1099	(NICAM), with a horizontal resolution of approximately 10 km or less in the target
1100	region, is proposed in the present study. Circulations over both the global and target
1101	domains are solved with a single model, whose mesh size varies with fine meshes
1102	covering the target region, to calculate meso-scale circulations in the study region. The
1103	stretched global model requires to computational costs to simulate atmospheric
1104	aerosols with fine horizontal resolutions compared with the global uniform
1105	nonhydrostatic modelt, whereas it may require higher computational costs compared
1106	with the general regional models, because the stretched-grid system calculates inside
1107	and outside the target domain. As opposed to the general regional models, the
1108	stretched-grid system does require neither nesting techniques nor boundary conditions.
1109	In this study, we developed the new <u>type regional</u> model with a horizontal resolution of
1110	approximately 10 km to simulate aerosols over Japan, especially in the megacities of the
1111	Kanto area, including Tokyo. To evaluate the model performances in the stretched-grid
1112	system (hereafter referred to as the "NICAM-g6str"), we also simulated
1113	NICAM-SPRINTARS with the globally uniformed grid simulation in glevel-6
1114	resolution (the horizontal resolution is set to 110 km and we call it "NICAM-g6"). Both
1115	NICAM-g6str and NICAM-g6 well reproduce general circulations obtained by
1116	reanalysis of NCEP-FNL under the nudging technique over Asia including the target

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-	Goto Daisuke 2014/11/22 21:04
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1124region. Only NICAM-g6str usually reproduces both diurnal and weekly vari1125the observed weather (temperature, wind, and precipitation) around Japa1126NICAM-g6str and NICAM-g6 generally reproduce monthly mean distribution1127observed sulfate and SO2 over East Asia, with the high correlations of more1128but the underestimation of the simulated concentrations by 40% (NICAM-ge)112950% (NICAM-g6). The underestimation is mainly caused by the underestim1130China and possibly by the uncertainty of the simulated precipitation around .1131the Kanto area, the results obtained by NICAM-g6. Only NICAM-g6str1133in simulating the wind patterns and the diurnal transitions around the center1134Kanto area, although it is inadequate to simulate the wind patterns and the1135transitions at some sites located at the edge of the Kanto area and surrounded1136sides by mountains, e.g., Maebashi, mainly due to the insufficient horizontal re1137NICAM-g6str also generally reproduces both diurnal and weekly variation
1126NICAM-g6str and NICAM-g6 generally reproduce monthly mean distribution1127observed sulfate and SO2 over East Asia, with the high correlations of more1128but the underestimation of the simulated concentrations by 40% (NICAM-gg)112950% (NICAM-g6). The underestimation is mainly caused by the underestim1130China and possibly by the uncertainty of the simulated precipitation around .1131the Kanto area, the results obtained by NICAM-g6str are much closer1132observations compared to those obtained by NICAM-g6. Only NICAM-g6str1133in simulating the wind patterns and the diurnal transitions around the center1134Kanto area, although it is inadequate to simulate the wind patterns and the1135transitions at some sites located at the edge of the Kanto area and surrounded1136sides by mountains, e.g., Maebashi, mainly due to the insufficient horizontal re
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1128 but the underestimation of the simulated concentrations by 40% (NICAM-g 1129 50% (NICAM-g6). The underestimation is mainly caused by the underestim 1130 China and possibly by the uncertainty of the simulated precipitation around . 1131 the Kanto area, the results obtained by NICAM-g6str are much closer 1132 observations compared to those obtained by NICAM-g6. Only NICAM-g6str 1133 in simulating the wind patterns and the diurnal transitions around the center 1134 Kanto area, although it is inadequate to simulate the wind patterns and the 1135 transitions at some sites located at the edge of the Kanto area and surrounded 1136 sides by mountains, e.g., Maebashi, mainly due to the insufficient horizontal results
112950% (NICAM-g6). The underestimation is mainly caused by the underestim1130China and possibly by the uncertainty of the simulated precipitation around 11131the Kanto area, the results obtained by NICAM-g6str are much closed1132observations compared to those obtained by NICAM-g6. Only NICAM-g6str1133in simulating the wind patterns and the diurnal transitions around the center1134Kanto area, although it is inadequate to simulate the wind patterns and the1135transitions at some sites located at the edge of the Kanto area and surrounded1136sides by mountains, e.g., Maebashi, mainly due to the insufficient horizontal results
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1133 in simulating the wind patterns and the diurnal transitions around the center 1134 Kanto area, although it is inadequate to simulate the wind patterns and the 1135 transitions at some sites located at the edge of the Kanto area and surrounded 1136 sides by mountains, e.g., Maebashi, mainly due to the insufficient horizontal results.
 1134 Kanto area, although it is inadequate to simulate the wind patterns and the 1135 transitions at some sites located at the edge of the Kanto area and surrounded 1136 sides by mountains, e.g., Maebashi, mainly due to the insufficient horizontal results
 1135 transitions at some sites located at the edge of the Kanto area and surrounded 1136 sides by mountains, e.g., Maebashi, mainly due to the insufficient horizontal results
1136 sides by mountains, e.g., Maebashi, mainly due to the insufficient horizontal re
1137 NICAM-g6str also generally reproduces both diurnal and weekly variation
1138 observed and/or a regional aerosol-transport model (WRF-CMAQ) simula
1139 sulfate, and SO_2 concentrations, especially with their high correlation (R
1140 Komae/Tokyo. The standard and sensitivity experiments suggest that (1)
1141 inventories of EC and SO_2 should be improved for the better simulation and (2)
1142 of the prescribed oxidants for the sulfate formation is not crucial for predicting
1143 and monthly averaged sulfate mass concentrations at least if the diurnal and
1144 variations of the prescribed oxidants are considered. As for PM2.5 simulation
1145 NICAM-g6str captures both weekly and diurnal cycles of PM2.5, with the exc

1146	the underestimation of the simulated PM2.5 by at least twice, probably due to the
1147	underestimation of secondary organic aerosol (SOA) from anthropogenic sources and
1148	the high uncertainties of the measurements,
1149	Therefore, this new seamless aerosol-transport model, which covers global to regional
1150	scales, can be applied to regional simulations. It suggests that even the simplified
1151	aerosol module (e.g., prescribed oxidants for sulfur chemistry) is applicable for the
1152	regional simulation if the module is coupled to a dynamic core with high horizontal
1153	resolution. To more accurately simulate areas around Japan and develop the simplified /
1154	aerosol module, we <u>need</u> to address the following objectives: (1) to increase the
1155	horizontal resolution (less than 10 km) to properly resolve wind fields, which can
1156	greatly influence the delivery of air pollution from Tokyo to subcities such as
1157	Maebashi; (2) to accurately reproduce the <u>cloud and precipitation fields</u> caused by
1158	thermal lows, for example, by applying the finer horizontal resolution and/or more
1159	sophisticated schemes of cloud microphysics such as the double-moment bulk scheme
1160	proposed by Seiki and Nakajima (2014); (3) to use better emission inventories by

1161	developing a data assimilation such as the Kalman smoother proposed by Schutgens et
1162	al. (2012) with intensive measurements in many sites; (4) to simulate strong peaks of
1163	PM2.5 in the daytime in the Kanto region by implementing more sophisticated module

1164	of SOA formed from both anthropogenic and biogenic sources, such as the volatility

1165 <u>basis-set approach proposed by Donahue et al. (2006), in this model; and (5) to treat</u>

1166 nitrate aerosol through a thermodynamic equilibrium in the simulation of wintertime

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削除: We discovered that this model can simulate
meteorological fields and anthropogenic primary
particles, e.g., elemental carbon (EC), and secondary
particles, e.g., sulfate, against in-situ measurements
and other regional models.

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daytime in the Kanto region

1187	of sulfate under the changes in emission of NO_x and SO_2 (e.g., Ohara et al., 2007).
1188	These issues are directly connected to the further development of NICAM-SPRINTARS
1189	in both regional and global simulations. Near the future, we will present scenario
1190	experiments at regional scales of 10 km grids and/or address the issue of regional air
1191	quality and its health impacts in densely populated megacities.
1192	
1193	Acknowledgements
1194	We acknowledge the developers of NICAM and SPRINTARS, especially K. Suzuki
1195	and T. Takemura, and the researchers from FAMIKA, especially S. Hasegawa and Y.
1196	Morino, and Y. R. Li and A. Miyaji for their assistance with processing the datasets. We
1197	are grateful to the GTOPO30 courtesy of the U.S. Geological Survey, the NCEP-FNL,
1198	EAGrid2000 by A. Kannari, and the local government measurements provided by the
1199	Tokyo Environment Agency, the Gunma Prefectural Institute of Public Health and
1200	Environmental Sciences and the Kawasaki Municipal Research Institute for
1201	Environmental Protection. We are also grateful to the working group members of
1202	Project SALSA, and the Ministry of Education, Culture, Sports and Science and
1203	Technology (MEXT), Some of the authors were supported by Project SALSA, which is
1204	part of the Research Program on Climate Change Adaptation (RECCA) by the MEXT

and/or future scenarios where the relative contribution of nitrate will be larger than that

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Goto Daisuke 2014/11/17 18:35
削除: has not been previously performed. The high
horizontal resolution can provide estimates of human
health impacts due to PM2.5. The findings from our
scenario experiment demonstrated the relevance of
estimating future concentrations, particularly in
aging populations with growing vulnerability. The
novel technique that combines the use of
Stretch-NICAM-SPRINTARS and pre-calculated
climate simulations by MIROC-AOGCM can
provide new opportunities to

削除: , who were responsible for the scenario experiments, Goto Daisuke 2014/11/8 15:21 削除: /Kakushin project, especially M. Watanabe, T. Nozawa and H. Kanai, for providing the MIROC datasets of climate simulation and emission

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inventories

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JAXA/EarthCARE, GCOM-C, MEXT/VL for

in Japan, the Global Environment Research Fund S-12 and A-1101 of the Ministry of

the Environment (MOE) in Japan, MOE/GOSAT, JST/CREST/EMS/TEEDDA,

climate

diagnostics

and

- 1228 MEXT/KAKENHI/Innovative Areas 2409. The model simulations were performed
- 1229 using supercomputer resources, SR16000 and PRIMEHPC FX10 from the University of
- 1230 Tokyo, Japan.

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1585	chemical signature, regional haze distribution and comparisons with global aerosols,
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1587 Table 1. Statistical values (<u>averages</u> of the observation and simulations, correlation

1588 coefficient R and root-mean-square-error RMSE) for meteorological fields using the

1589 simulations (NICAM-g6str and NICAM-g6) and observations at seven sites during the

same period, as shown in Figures 4 to 7.

1590

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

Goto Daisuke 2014/11/21 17:51 削除: mean Goto Daisuke 2014/11/8 14:40 削除: absolute bias *Ba*,

Goto Daisuke 2014/11/8 14:39

削除: Stretch-NICAM-SPRINTARS

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

1595

Table 2. Statistical values (averages of the observation and simulations, correlation 1596coefficient R and root-mean-square-error RMSE) for EC, sulfate, and SO2

1597

concentrations by the simulations (NICAM-g6str, NICAM-g6, and WRF-CMAQ) and 1598the observations at four FAMIKA sites during the period from August 6 to 11. The

1599WRF-CMAQ results are given by Shimadera et al. (2013).

		Maebashi	<u>Kisai</u>	Komae	<u>Tsukuba</u>
			Ē	<u>iC</u>	
Average $[\mu g/m^3]$	Observation	<u>2.85</u>	<u>2.75</u>	<u>1.23</u>	<u>2.20</u>
and difference [%]	NICAM-g6str	<u>0.39 (-86)</u>	<u>0.60 (-78)</u>	<u>1.10 (-10)</u>	<u>0.73 (-67)</u>
(vs. observation) in	NICAM-g6	<u>0.52 (-82)</u>	<u>0.52 (-81)</u>	<u>0.49 (-60)</u>	<u>0.58 (-74)</u>
<u>bracket</u>	WRF-CMAQ	<u>0.87 (-69)</u>	<u>1.17 (-58)</u>	<u>0.92 (-25)</u>	<u>0.77 (-65)</u>
<u>R</u>	NICAM-g6str	<u>-0.02</u>	<u>0.41</u>	<u>0.55</u>	<u>0.59</u>
	NICAM-g6	<u>-0.49</u>	<u>-0.28</u>	<u>-0.05</u>	<u>0.16</u>
	WRF-CMAQ	<u>0.08</u>	<u>0.33</u>	<u>0.37</u>	<u>-0.23</u>
<u>RMSE [µg/m³]</u>	NICAM-g6str	<u>2.62</u>	<u>2.33</u>	<u>0.72</u>	<u>1.85</u>
	NICAM-g6	<u>2.52</u>	<u>2.45</u>	<u>1.10</u>	<u>2.06</u>
	WRF-CMAQ	<u>2.18</u>	<u>1.83</u>	<u>0.88</u>	<u>1.98</u>
			<u>Sul</u>	fate	
Average [µg/m ³]	Observation	<u>4.79 (-6)</u>	<u>2.86 (44)</u>	<u>4.18 (-32)</u>	<u>4.85 (-12)</u>
and difference [%]	NICAM-g6str	<u>4.51 (-34)</u>	<u>4.14 (11)</u>	<u>2.84 (-46)</u>	<u>4.25 (-26)</u>
(vs. observation) in	NICAM-g6	<u>3.17 (-21%)</u>	<u>3.17 (42%)</u>	<u>2.25 (-21%)</u>	<u>3.58 (-22%)</u>
<u>bracket</u>	WRF-CMAQ	<u>3.77</u>	<u>4.08</u>	<u>3.30</u>	<u>3.80</u>

_					
<u>R</u>	NICAM-g6str	<u>0.01</u>	0.50	0.51	0.73
	NICAM-g6	0.05	<u>0.56</u>	0.86	<u>0.75</u>
	WRF-CMAQ	0.41	0.02	<u>0.87</u>	<u>0.78</u>
<u>RMSE [μg/m³]</u>	NICAM-g6str	<u>3.61</u>	<u>2.81</u>	<u>2.71</u>	<u>2.49</u>
	NICAM-g6	<u>3.01</u>	<u>2.30</u>	<u>2.49</u>	<u>2.77</u>
	WRF-CMAQ	2.30	<u>3.37</u>	<u>1.62</u>	<u>2.56</u>
			<u>S</u>	<u>O_</u>	
Average [ppbv]	Observation	2.74	<u>2.28</u>	<u>2.35</u>	<u>3.79</u>
and difference [%]	NICAM-g6str	<u>1.25 (-54)</u>	<u>1.90 (-17)</u>	<u>2.34 (-1)</u>	<u>2.34 (-38)</u>
(vs. observation) in bracket	<u>NICAM-g6</u>	<u>2.42 (-12)</u>	<u>2.45 (7)</u>	<u>2.52 (7)</u>	<u>3.21 (-15)</u>
R	NICAM-g6str	0.02	<u>-0.04</u>	0.62	<u>0.21</u>
	NICAM-g6	<u>-0.64</u>	<u>-0.52</u>	<u>0.22</u>	<u>-0.04</u>
RMSE [ppbv]	NICAM-g6str	<u>1.82</u>	<u>0.93</u>	<u>0.97</u>	<u>2.08</u>
	NICAM-g6	<u>1.29</u>	<u>0.94</u>	<u>0.85</u>	<u>1.29</u>

1601 Table 3. PM2.5 concentrations in daily, daytime (from 9 am to 4 pm), and nighttime

1602 (from 9 pm to 4 am) averages and mean ratios of daytime to nighttime using the

1603 simulations (NICAM-g6str and NICAM-g6) and the observation at selected seven sites

1604

in August.								
	<u>Maebashi</u>	<u>Kawasaki</u>	<u>Toride</u>	<u>Hasuda</u>	<u>Sapporo</u>	<u>Nagoya</u>	<u>Fukuoka</u>	
		Daily mean	PM2.5 [µg/	m ³] and stan	dard deviati	on [µg/m ³]		
Observation	<u>24.9±12.8</u>	<u>23.2±12.9</u>	<u>17.6±9.7</u>	<u>20.6±11.5</u>	<u>12.7±6.3</u>	<u>17.3±10.1</u>	<u>14.3±7.5</u>	
NICAM-g6str	<u>6.4±3.9</u>	<u>10.0±7.3</u>	<u>9.0±6.3</u>	<u>8.4±5.0</u>	<u>4.9±3.5</u>	<u>7.5±5.7</u>	<u>3.4±2.6</u>	
NICAM-g6	<u>6.7±3.0</u>	<u>6.7±3.3</u>	<u>6.7±3.4</u>	<u>6.7±3.0</u>	<u>4.7±4.1</u>	<u>5.4±3.0</u>	<u>3.5±2.3</u>	
	<u>Daytim</u>	e (9am-4pm)) mean PM2	2.5 [µg/m ³] a	nd standard	deviation [µ	g/m ³]	
Observation	<u>28.6±14.1</u>	<u>19.4±12.1</u>	<u>15.8±9.0</u>	<u>21.0±10.0</u>	<u>15.0±5.2</u>	<u>11.3±5.4</u>	<u>9.7±5.7</u>	
NICAM-g6str	<u>5.9±3.8</u>	<u>7.1±4.3</u>	<u>6.8±4.4</u>	<u>7.2±4.5</u>	<u>5.3±2.8</u>	<u>3.5±2.3</u>	<u>1.6±0.8</u>	
NICAM-g6	<u>5.0±1.7</u>	<u>4.0±2.1</u>	<u>4.0±2.4</u>	<u>4.4±1.9</u>	<u>7.4±4.5</u>	<u>2.4±0.9</u>	<u>1.4±0.5</u>	
	Nighttime (9pm-4am) mean PM2.5 [$\mu g/m^3$] and standard deviation [$\mu g/m^3$]							
Observation	<u>24.4±11.9</u>	<u>24.5±11.8</u>	<u>16.9±9.6</u>	<u>18.5±10.3</u>	<u>10.7±6.6</u>	<u>19.1±8.2</u>	<u>15.4±6.7</u>	
NICAM-g6str	<u>7.5±3.6</u>	<u>14.2±9.2</u>	<u>12.1±7.6</u>	<u>10.8±5.5</u>	<u>4.1±3.9</u>	<u>12.0±4.6</u>	<u>5.1±3.1</u>	

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

1606	Figure captions	
1607	Figure 1 Topographical maps of (a) East Asia and (b) Eastern Japan , including the	Goto Daisuke 2014/11/8 15:22 削除: Japan
1608	observation sites for the model validation, The topography is based on GTOPO30 (the	Goto Daisuke 2014/11/16 13:06
1609	horizontal resolution is 30 arc seconds, that is approximately 1 km) courtesy of the U.S.	削除: the Kanto Goto Daisuke 2014/11/16 13:06
		削除: region
1610	Geological Survey,	Goto Daisuke 2014/11/18 17:25
1611		削除: of the model
1612	Figure 2 (a) EC and (b) SO ₂ emission inventories in 2007,	Goto Daisuke 2014/11/8 15:23 削除: The circles represent eight sites (1.
1613		Maebashi/Gunma, 2. Kisai/Saitama, 3.
1015		Komae/Tokyo, 4. Tsukuba/Ibaraki, 5.
1614	Figure 3 Horizontal distributions of temperature and winds in August averages at the	Yokohama/Kanagawa, 6. Chiba/Chiba, 7.
1615	surface and the model height of approximately 5 km for the model bottom of MSL over	Adachi/Tokyo and 8. Machida/Tokyo). Goto Daisuke 2014/11/8 15:24
1616	Asia region using reanalysis data from NCEP-FNL, simulation by NICAM-g6str, and	削除: from
1617	simulation by NICAM-g6,	Goto Daisuke 2014/11/8 15:24 削除: (a)
1618		Goto Daisuke 2014/11/8 15:24
1010		削除: for the standard experiment and (b) 2030 for
1619	Figure 4 Temporal variations in the NICAM-g6str and NICAM-g6 simulated and	the RCP4.5 scenario experiment
1620	observed air temperature for a height of 2 m at (a) Yokohama, (b) Chiba, (c) Tsuchiura,	Goto Daisuke 2014/11/8 15:25 削除: Same as Figure 2 but for SO ₂ emission
1621	(d) Adachi, (e) Maebashi, (f) Machida and (g) Kisai in August 2007.	inventories
		Goto Daisuke 2014/11/16 13:08
1622		削除: Stretch-
1623	Figure 5 Same as Figure 4 but for relative humidity (RH).	Goto Daisuke 2014/11/8 15:29
1 00 1		削除: from
1624		Goto Daisuke 2014/11/8 15:29 削除: 4-24,
1625	Figure 6 Same as Figure 4 but for wind direction.	Goto Daisuke 2014/11/16 13:08
1626		削除: The numbers located in the upper right corner
		of each panel show the simulated and observed mean
1627	Figure 7 Same as Figure 4 but for wind speed.	values.

1649			
1650	Figure <u>8 Horizontal distributions of precipitation in August averages</u> derived from (a)		Goto Daisuke 2014/11/8 15:27
1651	simulation by NICAM-g6str, (b) simulation by NICAM-g6, (c) reanalysis data from		下へ移動 [3]: Figure 8 Temporal variation in the
1652	MSM by JMA and (d) reanalysis data from GSMaP.		simulated Stretch-NICAM-SPRINTARS and observed precipitation amounts at (a) Yokohama, (I
1653			Chiba, (c) Tsukuba, (d) Koutou, (e) Maebashi, (f) Huchu and (g) Konosu from August 4–24, 2007.
1654	Figure 9 Temporal variations in the NICAM-g6str_and NICAM-g6 simulated and		Goto Daisuke 2014/11/16 13:09
1655	observed precipitation amounts at 21 Japanese sites in August 2007. The comparison		削除: Figure 8 Temporal variation in the simulated simulated Stretch-NICAM-SPRINTARS and
1656	includes 10 sites in the Kanto area; (a) Maebashi, (b) Konosu, (c) Huchu, (d) Tsukuba,		observed precipitation amounts at (a) Yokohama, (I
1657	(e) Tokyo, (f) Yokohama, (g) Abiko, (h) Saitama, (i) Chiba, and (j) Nerima, 3 sites in		Chiba, (c) Tsukuba, (d) Koutou, (e) Maebashi, (f) Huchu and (g) Konosu from August 4–24, 2007.
1658	the northern Japan; (k) Niigata, (l) Sendai, and (m) Sapporo, 5 sites in the western		Goto Daisuke 2014/11/8 15:27 移動 (挿入) [3]
1659	Japan; (n) Nagoya, (o) Osaka, (p) Himeji, (q) Fukuoka, and (r) Hyuga, and 3 remote		Goto Daisuke 2014/11/8 15:27 削除: 8 Temporal variations in the simulated
1660	islands (s) Hachijo-jima, (t) Oshima, and (u) Naha.		
1661			
1662	Figure 10 Predictive values of daily precipitation using the NICAM-g6str and		
1663	NICAM-g6 simulations and the AMeDAS measurements during August 2007 at the		
1664	sites defined at Figure 9, in units of percentage.		
1665			
1666	Figure <u>11</u> Temporal variations in <u>the simulated</u> (NICAM-g6str, NICAM-g6, and		Goto Daisuke 2014/11/8 15:30
1667	WRF-CMAQ) and observed EC mass concentrations, near the surface at (a) Maebashi,	/	削除: 101 Temporal variations in
1668	(b) Kisai, (c) Komae and (d) Tsukuba <u>in</u> August 2007, <u>The WRF-CMAQ results are</u>		
1669	given by Shimadera et al. (2013). The left axis in red represents the simulated values,		
1670	and the right axis in black represents the observed values, in units of $\mu g/m^3$.		

(g) Konosu from August 4–24, 2007.

... [15]

ecipitation amounts at (a) Yokohama, (b) sukuba, (d) Koutou, (e) Maebashi, (f) (g) Konosu from August 4-24, 2007. uke 2014/11/16 13:09 re 8 Temporal variation in the simulated retch-NICAM-SPRINTARS and ecipitation amounts at (a) Yokohama, (b) sukuba, (d) Koutou, (e) Maebashi, (f)

1716			
1717	Figure <u>12</u> Same as Figure <u>11</u> but for sulfate.		Goto Daisuke 2014/11/8 15:30
1718			削除: 112 Same as Figure 10
1719	Figure <u>13</u> Same as Figure <u>12</u> but for SO ₂ , without the WRF-CMAQ results, in units of		Goto Daisuke 2014/11/8 15:31
1720	ppbv.		削除: 123 Same as Figure 102 but for SO,
1721			
1722	Figure <u>14 Scatterplot</u> of <u>August mean</u> concentrations <u>for EC</u> , sulfate and SO ₂ between		Goto Daisuke 2014/11/8 15:31
1723	the simulations by NICAM-g6str and NICAM-g6 and the observations at the sites		削除: 134 Comparisoncatterplot of August
1724	shown in <u>the left</u> panels,		
1725			
1726	Figure <u>15 Horizontal distributions of</u> concentrations for <u>EC</u> , sulfate and SO_2 near the		Goto Daisuke 2014/11/8 15:32
1727	surface using NICAM-g6str and NICAM-g6 in August averages. The circles in color		削除: 145 Horizontal distributions of Average
1728	shows the observation results at the sites.		
1729			
1730	Figure <u>16 (a)</u> EC and (b) sulfate mass concentrations at the FAMIKA <u>four</u> sites using		Goto Daisuke 2014/11/8 15:33
1731	NICAM-g6str under the sensitivity experiments, WRF-CMAQ results shown by	\square	削除: ,
1732	Shimadera et al. (2013) and the FAMIKA observations in averages of August 6-11. The	//	
1733	bar represents the range of the sensitivity,		
1734			
1735	Figure <u>17</u> Temporal <u>variations</u> in <u>the NICAM-g6str and NICAM-g6</u> simulated and		Goto Daisuke 2014/11/8 15:35
1736	observed PM2.5 near the surface at <u>18 Japanese sites in</u> August 2007. The left axis in		削除: 167 Temporal variationariations in the
1737	red represents the simulated values, and the right axis in black represents the observed		

1785	values, in unit of μ gm ⁻³ .	Goto Daisuke 2014/11/8 15:39
1786		削除: The numbers located in the upper right corner of each panel show the simulated and observed mean
1787	Figure <u>18 Extinction coefficients in August averages</u> for the spherical particles	values.
1788	simulated by <u>NICAM-g6str, and NICAM-g6</u> and the spherical particles observed by the	Goto Daisuke 2014/11/8 15:35 削除: 17
1789	NIES-LIDAR network, at (a) Tsukuba and (b) Chiba, in units of 1/(Mm), The bars	Goto Daisuke 2014/11/18 18:02
1790	represent the 25th and 75th percentiles of the LIDAR observations.	削除: Average e Goto Daisuke 2014/11/8 15:39
		削除: Stretch-NICAM-SPRINTARS
		Goto Dajeuka 2011/11/8 15:30

削除: (shown in red)

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