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Interactive comment on "Application of a global nonhydrostatic model with a stretched-grid system to regional aerosol simulations around Japan" *by* D. Goto et al.

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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

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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 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. 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 mass concentration obtained 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. 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 large-scale atmospheric sulphate aerosol models (COSAM): overview and highlights, Tellus, 53B, 615-645, 2001. Donahue, N. M., Robinson, A. L., Stanier, C. O., and Pandis, S. N.: Coupled partitioning, dilution, and chemical aging of semivolatile organics, Environ. Sci. Technol., 40, 2635-2643, 2006. Hallquist, M., Wenger, J. C., Baltensperger, U., Rudich, Y., Simp- son, D., Claeys, M., Dommen, J., Donahue, N. M., George, C., Goldstein, A. H., Hamilton, J. F., Herrmann, H., Hoff- mann, T., Iinuma, Y., Jang, M., Jenkin, M. E., Jimenez, J. L., Kiendler-Scharr, A., Maenhaut, W., McFiggans, G., Mentel, Th. F., Monod, A., Prevot, A. S. H., Seinfeld, J. H., Surratt, J. D., Szmigielski, R., and Wildt, J.: The formation, properties and impact of secondary organic aerosol: current and emerging issues, Atmos. Chem. Phys., 9, 5155–5236, doi:10.5194/acp-9-5155- 2009, 2009. 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. Kajino, M., and Kondo, Y.: EMTACS: Development and regional-scale simulation of a size, chemical, mixing type, and soot shapre resolved atmospheric particle model, J. Geophys. Res., 116, D02303, doi:10.1029/2010JD015030, 2011. Shimadera, H., Hayami, H., Chatani, S., Morino, Y., Mori, Y., Morikawa, T., Yamaji, K., and Ohara, T.: Sensitivity analyses of factors influencing CMAQ performance for fine particulate nitrate, J. Air and Waste Manag. Assoc., 64(3), 374-384, 2014.

[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-

GMDD

7, C2425–C2438, 2014

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 SO2 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 SO2 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

GMDD
7, C2425–C2438, 2014

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."

[Reference] 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.

GMDD

7, C2425–C2438, 2014

[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

GMDD
7, C2425–C2438, 2014

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 pollu-

GMDD
7, C2425–C2438, 2014

tion 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

GMDD

7, C2425–C2438, 2014

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-

GMDD

7, C2425–C2438, 2014

SPRINTARS simulations?

[A2-12] Although in the original manuscript, the spin-up time was just one week, we re-calculated 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 over-predicts 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

GMDD

7, C2425–C2438, 2014

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.

GMDD
7, C2425–C2438, 2014

Interactive comment on Geosci. Model Dev. Discuss., 7, 131, 2014.

GMDD

7, C2425–C2438, 2014