Title: Aerosol dynamics and gas-particle conversion in dry deposition of inorganic reactive nitrogen in a temperate forest

Authors: G. Katata et al.

Author response to reviewer comments

Response to Anonymous Referee #2

The paper presents a 1-dimensional model of coupled phase-partitioning and multi-layer canopy to study the process of NH4NO3 aerosol evaporation and water uptake on surface atmosphere exchange fluxes of reactive nitrogen compounds. A number of studies have observed remarkably high apparent deposition rates of ammonium and nitrate aerosol which have been linked to aerosol evaporation processes, but few studies have attempted to model the process and thereby quantify the overall effect on N deposition. This paper presents a detailed model for this purpose and applies to reproduce concentration gradients measured above a Japanese forest. Whilst the model is detailed, the measurements are comparably basic and poorly time-resolved which limits their value in really testing the model. Nevertheless, the paper presents a useful first application of the model and makes a valuable contribution to the literature on the subject. I recommend its publication once a few concerns have been addressed as detailed below.

Response: We sincerely appreciate your interests and positive comments on our manuscript. In the original manuscript, we should discuss about the model results via the uncertainties of measurements more carefully as a first application (not for model validation). Therefore, as responded below we revised the whole sentences throughout the manuscript regarding the input data based on your comments and suggestions. We hope the manuscript should be clearly improved. For example, we inserted the sentences about the uncertainty of the datasets: ”In this study, we made a simulation with basic and less time-resolved datasets as very first application of the model to the NH4NO3 gas-particle conversion and aerosol water uptake of reactive nitrogen compounds. The above uncertainties associated with input data such as number concentration and particle size distribution should be improved in future.” (L.239-241, p.8).

Importantly, the description of the model and its application is very brief and important detail is missing. In addition, the authors could exploit their results a little more comprehensively. My suspicion is that this would result in a sufficient amount of new, additional text to necessitate a re-review of the manuscript.

Response: A part of model description and simulation conditions were not enough for evaluation. As responded to your suggestions and comments below, the manuscript was drastically improved. Thank you to your comments.

Major concern

1. My major concern relates to the use of long-term average concentrations. Please make the time resolution of

the filter pack measurements more explicit. It suggests in Line 147 that integration times were 9 hrs (day-time) and 15 hrs (night-time), whilst line 284 talks about weekly measurements. In either case I do not understand why they were used as spot measurements in the interpolation (Figs. 1 and 4). Should the model not be initialised with the same concentration for those time-intervals? Otherwise the average of the interpolated time-series does not match the measured average.

Response: The description about filter-pack sampling was confusing and caused misunderstanding of reviewer. For the early autumn period, filter-pack sampling was continuously made during the day and night for no rain days. For the late autumn period, the time resolution was relatively low as weekly continuous measurements. However, the data in rain days of the early autumn period were missing and caused gaps as shown in old Figs. 1 and 4. As a result, we had no choice to make the data of these gap periods linearly incorporated for simulations. As you suggested, since this interpolation could cause unrealistic effects on our results and should not be analyzed, we shaded the output data during the rain period (no available filter-pack data) in new Fig. 6. In the original manuscript, this error should not affect our results (old Fig. 5) because we used only the calculations and measurements in the above no rain period for comparisons of inorganic mass concentration. Regarding initialization, 12 hour was applied to spin-up time in this study, which is considered to be sufficiently low the gas and particle exchanges between atmosphere and vegetation was sufficiently higher than time resolution of filter-pack data (~ half day). To address this, we revised the sentences as ”For the early autumn period, filter-pack sampling was continuously performed during the day and night except when it was raining. As a result, 5 daytime reading data sets and 6 nighttime reading data sets were available. The gaps between data in rain days of the early autumn period were linearly incorporated for simulations. Since the interpolation could cause unrealistic effects on the results, we used only the calculations and measurements in the above no rain period for comparisons of inorganic mass concentration. For the late autumn period, the time resolution was relatively low as weekly continuous measurements.” (L.172-177, p.6) and “The model profiles averaged for only the sampling periods were compared with observed ones.” (L.276, p.9).

Similarly, the length of the intervals of the dots in Figure 4 do not suggest that the measurements were continuous. Line 251 and Figure 6. I would be very surprised if the conditions for NH4NO3 condensation were met in this forest. Normally, these conditions are found over strong sources of NH3 (e.g. Nemitz et al., 2009). If I understand the manuscript correctly, the authors are using long-term average concentration measurements with long-term average meteorology. In general, the vapour pressures of NH3 and HNO3 are dependent on temperature and humidity in a highly non-linear relationship. This means, if long-term average concentrations are paired with long-term averages in temperature and humidity, it is unlikely that the comparison of measured concentrations with the thermodynamic equilibrium concentrations evaluated with the ISORROPIA2 model can correctly assess whether there is potential for NH4NO3 evaporation of condensation. With the long-term integrated samples this problem cannot be fully resolved and as a result the entire manuscript needs to be reformulated to some degree that full agreement between modelled and measured conditions cannot be expected. The application to this measurement dataset can only be considered a first test of the model, rather

than a conclusive assessment of its capability. This problem needs to be discussed and, potentially, a sensitivity analysis could explore the uncertainty introduced by the averaging. In addition, the concentrations used for initialisation are subject to measurement uncertainty that may further limit the model / measurement comparison.

Response: Again, our description was unclear and caused misunderstanding of the reviewer. The input filter-pack data of was half-daily with linear interpolation (early autumn period) or weekly (late autumn period), but the output data of all mass and number concentrations was half-hourly as well as all meteorological variables (old Fig. 3). These were averaged for only the sampling periods for comparisons with observations. Therefore, the effect of diurnal changes of air temperature and humidity should be reflected to the results in the early autumn period, which is not so problematic, we believe. In order to emphasize that this work is a first application of the model, the details in uncertainties are added based on your suggestions: "Several uncertainties (e.g., low time resolution of weekly filter-pack data in the late autumn period; initialization of measurement uncertainty; complex topography of the study site) may cause underestimations in calculated wind speed (Fig. 3a and 4a) and overestimations in total number concentration within the canopy after 25 November 2016 (Fig. 9a). In Fig. 8, the conditions for NH4NO3 condensation were calculated in this forest, although these conditions are normally found over strong sources of NH3 (e.g. Nemitz et al., 2009). Thus, the results from this study can only be considered a first test of the model to the NH4NO3 gas-particle conversion and aerosol water uptake of reactive nitrogen compounds, rather than a conclusive assessment of its capability." In addition, the name of subsection was also revised as "Uncertainties in observation and model results". (L.324-331, p.11).

2. Overall, I am missing results and discussion on the effect of gpc (and also the equilibration with water content) on the exchange with the vegetation, in addition to the effect on the fluxes above the canopy. Only if this effect is shown to be significant, would there a need to incorporate this additional complexity into deposition schemes. To assess the importance, I would encourage the authors to quantify, from their results, the effect on the actual ecosystem flux of the various forms of N, total reactive N and also the effective bulk deposition velocity of the aerosol at the surface (V_{ds}), which changes because particle size changes. Presumably, this change in V_{ds} is the reason for gpc changing also the in-canopy gradient of SO_4^{2-} (Fig. 5) although it does not take part in the gpc process itself. The implications should be discussed.

Response: Thank you so much for your crucial suggestions. We calculate the changes in apparent mass flux over the canopy and the contribution of each compounds on total nitrogen flux. We also showed the vertical profiles of mass flux as well as concentration (Fig. S2). To demonstrate the impact of gpc on particle (especially NO3-) deposition flux, the following discussion was inserted: "Once the gpc process is considered, particle deposition could have very important contribution as nitrogen flux over the forest ecosystem. Comparing the calculated daytime mass flux at 30 m height between "no gpc" and "gpc" scenarios in the early autumn period (Fig. S2),

deposition flux of fine NO3- and NH4+ was 15 and 4 times higher in "gpc" scenario. Since there was almost no change in SO42- flux between two scenarios, this change is only caused by gpc. For gas species, both HNO3 and NH3 slightly decreased to 0.6 and 0.8 times due to evaporation of NH4NO3 particles. This change in flux could be applied to that in deposition velocity of each species. Furthermore, although particle deposition flux contributes to only 5 % of total nitrogen flux above the canopy in "no gpc" scenario, this impact was increased to 38 % (NO3-: 27 %, NH4+: 11 %) in "gpc" scenario. It should be noted that the contribution of NH3 was still large as 37 % in total nitrogen flux even in the "gpc" scenario. The above results indicate that the increase of (apparent) particle deposition due to NH4NO3 evaporation may be important in the chemical transport modeling.". (L.391-400, p.13). Part of this explanation also appeared in Abstract.

3. Related to this, the discussion of Figure 9 is very cryptic and only accessible to those already very familiar with the subject. It is closely linked to the observations of apparent bi-directional size-segregated fluxes (e.g. Nemitz and Sutton, 2004; Ryder, 2010) and this link should be made.

Response: Thank you for introducing Ryder (2010). We knew the paper from Nemitz (2015), but we did not refer it because we could not have an access to the document as it is the doctoral thesis. As you suggested, we incorporated additional explanation as "This is a similar result to demonstrated by past numerical study for size-resolved particle number flux (Ryder, 2010); the certain particle diameter around 0.15 μm at which the apparent flux switched from deposition to emission within the canopy and approximately reflected the peak in the number size distribution. Furthermore, the apparent emission flux was represented as more particles shrink into a given size bin from the next larger size than are leaving the bin to the next smaller size, while more particles shrink out of a given size bin than shrink into it from the next larger size bin, resulting in apparent fast deposition (Ryder, 2010)." (L.309-314, p.10-11).

Additional scientific comments and needs for clarification:

○Abstract. It would be helpful to be more quantitative and also to include a statement on the effect on the NH3 flux as well as the total reactive N flux. By what fraction does the change in phase partitioning change the net N flux?

Response: Thank you for your suggestion. We calcualted the contribution of all reactive nitrogen components (including NH3) to total nitrogen flux as "Once the gpc process is considered, particle deposition could have very important contribution as nitrogen flux over the forest ecosystem. Comparing the calculated daytime mass flux at 30 m height between "no gpc" and "gpc" scenarios in the early autumn period (Fig. S2), deposition flux of fine NO3- and NH4+ was 15 and 4 times higher in "gpc" scenario. Since there was almost no change in SO42- flux between two scenarios, this change is only caused by gpc. For gas species, both HNO3 and NH3 slightly decreased to 0.6 and 0.8 times due to evaporation of NH4NO3 particles. This change in flux could be applied to that in

deposition velocity of each species. Furthermore, although particle deposition flux contributes to only 5 % of total nitrogen flux above the canopy in "no gpc" scenario, this impact was increased to 38 % (NO3-: 27 %, NH4+: 11 %) in "gpc" scenario. It should be noted that the contribution of NH3 was still large as 37 % in total nitrogen flux even in the "gpc" scenario. The above results indicate that the increase of (apparent) particle deposition due to NH4NO3 evaporation may be important in the chemical transport modeling." (L. 391-400, p.13). Part of this explanation also appeared in Abstract.

○Line 65. Please clarify if the model also predicts the relative humidity profile which is important in controlling the phase partitioning. Related to this, Figure 3 should convey better whether the measured (in-canopy) profiles in the meteorological parameters are correctly reproduced by the model, i.e. the emphasis should be on the vertical change rather than the time-series.

Response: Since the relative humidity profile was predicted in the atmosphere sub-model, we added the word "relative humidity" as well as specific humidity at the line 63 in p.3. Furthermore, as you suggested in your main concern, we should emphasize that this was not model validation study but very first application to using limited datasets. Thus, this study should not demonstrate agreement between calculations and observations (as performance test), so that we should much focus on comparisons as first application of the model to the NH4NO3 gas-particle conversion. As a result, we added new Fig. S1 for vertical profiles of meteorological variables and revised the sentences as "For calculated air temperature and humidity, the primary determinants of ambient conditions of gas-particle conversion and aerosol hygroscopic growth, calculated temporal changes were closed to the observations within the canopy (Fig. 3 and 4; b and c). The above features were found in mean vertical profiles during the daytime and nighttime (Fig. S1)." (L.258-261, p.9).

○Lines 100ff. I do not understand the approach taken for calculating χd. The text reads as if this is chosen to match the atmospheric concentration at each canopy layer. Surely, in this case Fgd becomes zero if the canopy layer air concentration matches the gas phase concentration in equilibrium with the Γd of the leaf water layer, and rd ceases to have any effect. It is exactly the departure from equilibrium that drives the flux. Instead, Γd is controlled by the previous accumulated deposition onto the leaf cuticle and the size of the water pool. How is the water pool size calculated in the model? Related to this, I am not convinced the use of an rd that is linked to acid/NH3 ratio and a leaf water emission potential are internally consistent. Parameterisations of the cuticular resistance as a function of this ratio have been developed and applied within the framework of a zero leaf water emission potential (e.g. Nemitz, 2015; Fowler et al., 2009) meant to account for the effect of a non-zero leaf water concentration. By additionally introducing a non-zero Γd value, this effect is accounted for twice.

Response: The model description related to χd for wet canopy was not enough. We incorporate the sentences how to calculate leaf surface water from Katata et al. (2013) as follows: "The RH value could be affected by the leaf surface water content predicted at each canopy layer, based on water balance due to evaporation of leaf surface

water, interception of precipitation by leaves, capture of fog water by the leaves, and the drip from leaves (Katata et al., 2008; 2013)." (L.118-121, p.4-5). The surface water capacity of 0.2 kg m-2 was given as leaf surface water amount where there is maximum evaporation by Deardorff (1978). The NH_3 exchange processes scheme has been validated at rice field in Japan (Katata et al., 2013), but as you suggested, there is the uncertainty for applying rd to the condition of non-zero leaf water concentration of NH_3. Thus, we added the sentences as "Since our model is not the dynamic modeling approach (e.g., Sutton et al., 1998; Flechard et al., 1999) to simulate NH_3 charging and discharging of the cuticle, Equation (4) could have uncertainty at wet canopy in equilibrium with non-zero leaf surface concentration of NH_3." (L.121-123, p.5).

Deardorff, J.W., 1978. Efficient prediction of ground surface temperature and moisture, with inclusion of a layer of vegetation. J. Geophys. Res. 83, 1889–1903.

○Line 113. I realise that the authors are here only summarising the principles of a paper that is described in more detail elsewhere. Nevertheless, it would help to cite the approach taken to estimate the aerosol capture efficiency. Similarly, the origin of Eq. (4) needs to be mentioned.

Response: We added the original references for Eqs. (4) (Massad et al., 2010) and (6) (Fuchs, 1964; Kirsch and Fuchs, 1968; Peters and Eiden, 1992; Petroff et al., 2009).

○Line 130. In addition to the ISORROPIA2 thermodynamic module, does the model treat any gas-phase chemistry? This may be important as an additional source for HNO3. If not, the authors should discuss the implications somewhere.

Response: The gas-phase chemistry such as HNO3 production from NO_2, HONO, N_2O_5, and so on was not considered in the model. Since the process could cause uncertainties in our results as you suggested, we added the sentence as "The gas-phase chemical production of HNO_3 could affect the simulated HNO_3 concentration and flux, which should be implemented to the model in future." (L.153-154, p.6).

○Line 198. It is not clear how the leaf water content was prescribed. This should affect the overall RH profile throughout the canopy and thus the results should be quite sensitive to this parameter?

Response: Since the description about leaf water content calculation was missing in the manuscript, we added the sentences "The RH value could be affected by the leaf surface water content predicted at each canopy layer, water balance due to evaporation of leaf surface water, interception of precipitation by leaves, capture of fog water by the leaves, and the drip from leaves. (Katata et al., 2008; 2013)." (L. 118-121, p.4-5). This could increase the RH value in the canopy layer, although test periods for comparisons between observations and calculations were without rainfall (L.175-176, p.6).

○Line 203. The meaning of fio is not clearly introduced. I understand it to be the ratio of inorganic to total aerosol mass. If so, it is constrained by the observations (in contradiction to what is stated in the manuscript) by the comparison of total inorganic aerosol mass (from the filter-pack measurements) to total aerosol mass (approximated via the ELPI+ aerosol volume). What is not constraint is its size dependence.

Response: The description about fio estimation was confusing and further information about ELPI+ data was required. Your understanding about calculating fio for fine and Aitken modes is correct, but we required to assume its values for both fine and Aitken modes because the ELPI+ data (total mass concentration) was limited for only the late autumn period. Since the sentences were not organized, we revised them as follows: "In order to simulate the vertical profiles of total number concentration within the canopy, the volume fraction of inorganic compounds, fio, was given by the data of total inorganic and total mass concentrations. For the late autumn period, temporal changes in weekly fio values for fine and Aitken modes was given by the data of filter-pack and ELPI+ measurements. However, for the early autumn period, the ELPI+ measurements were unavailable as described in last subsection. Therefore, temporal changes in fio (Fig. 1b) were set based on the filter-pack data at the study site and total PM2.5 mass concentrations observed at the nearest air quality monitoring station at Hachiouji (3 km west-north-west from the site). For both periods, fio for Aitken mode was assumed to be the same as that for fine mode, since again no observational data were available." (L.229-236, p.8).

○Section 3.2. I am missing more explanation as to how the measurements were used to drive the model and provide initial or boundary conditions. In fact the content of this section does not match its title. Did you use concentrations at a single height (if so, which one?) or several heights? How often was the model re-initialised with the measurements. Was it allowed to run to steady-state conditions or was it continuously perturbed by the measurements? Was a spin-up time used? Maybe, the input could be illustrated by adding the constraining concentrations as a top panel to Figure 4.

Response: We should show detailed explanation about initial and boundary conditions. To present it more clearly, new Table 1 was inserted for both periods. No spin-up time was applied in this study because the gas and particle exchanges between atmosphere and vegetation was sufficiently higher than time resolution of filter-pack data (~ half day).

○In addition, it is numerically problematic to use the same aerosol composition across all sizes as the Kelvin effect then causes evaporation from the smallest particles and condensation on the larger ones. This would drive some of the changes in the diameters (Fig. 7), which would then not represent a response to vertical gradients but reflect inadequate initial conditions. How was this problem dealt with? In this context (and in general) I would encourage the authors to study and refer the work of Ryder (2010) who also developed a similar model and applied it to existing datasets of exchange.

Response: Since we are using a modal aerosol dynamics method, differences in Kelvin effect for fine and Aitken

mode particles are considered by using their mass mean diameter to represent the effect, but those within a mode cannot be resolved. This should be problematic as the standard deviation of the size distribution is enhanced. We somehow dealt with it by applying mode merging (or mode renaming), a part of Aitken mode (or accumulation mode) shifts to accumulation mode (or Aitken mode) when condensation (swelling) occurred to avoid unrealistically broad size distribution (e.g. Kajino et al., 2012). Certainly, our assumption, same composition in size at the initial and boundary conditions might cause large uncertainty in simulated vertical size distribution profile and its perturbation, but the size-resolved measurements have not been available so far. In future, we need to conduct size-resolved composition measurements to assess this uncertainty by using size-resolved numerical model (i.e., bin model).

Thank you for introducing Ryder (2010). We knew the paper from Nemitz (2015), but we did not refer it because we could not have an access to the document as it is the doctoral thesis. Consequently, in the revised manuscript, we added the following statements in subsection 5.1 with referring Ryder (2010): "Another uncertainty in the results could be associated with same composition assumption in size at the initial and boundary conditions. Variations of chemical composition in size caused variations in equilibrium vapor pressure at particle surface due to Kelvin and Raoult's effects, which caused uncertainty in the simulation of swelling and shrinking of particles. Since we used a modal aerosol dynamics method, differences of these effects within each mode cannot be resolved. We need to revisit this issue in the future by using size-resolved composition measurements and size-resolved aerosol model as done by Ryder (2010) to assess this uncertainty." (L.332-337, p.11).

○Line 210. Values of cg and cs as high as 300 ppb and 2000 ppb are completely unrealistic and inconsistent with the paper of Massad et al. (2010). They also ignore the temperature dependence of Eq. (3). Do the authors mean Gg = 300 and Gs = 2000 (no units!)? Gg is mentioned for the first time here and needs to be introduced much earlier on. Again, this calls for introducing the model in more detail than is currently done.

Response: The variables χg and χs were wrong and confusing the reviewer. These should be Γg and Γs (without unit), respectively. We modified the above variables and introduced them at new Eqs. (5) and (6), and modified.

○Section 5.1 and Line 349ff. Figure 4 suggests to me that the extrapolation of concentration into the canopy led to better agreement in the run that did NOT include gpc, whilst the text talks about an improvement. Please clarify. In general, to assess model performance, it would be much more illustrative to rearrange Figure 5 so that the three lines (Obs, no gpc, gpc) can be compared on a single plot for each compound. In addition, it would be interesting to compare a plot of how fluxes changes with height.

Response: As you suggested in your main concern, we should emphasize that this was not model validation study but very first application to using limited datasets. Thus, this study should not demonstrate model improvement and performance test, so that we should much focus on comparisons as first application of the model to the NH4NO3 gas-particle conversion. The sentences in Conclusions were now revised as "In the simulation including NH4NO3 gas-particle conversion processes, vertical gradients of normalized mass concentrations of nitrogen gases (HNO3

and NH3) and fine particles (NO3- and NH4+) within the canopy were clearly high than that for SO42-." (L.418-420, p.14). In section 5.2, the profiles of daytime flux were introduced as new Fig. S2 for all inorganic nitrogen components as "The above feature was also apparent in vertical profiles of mass flux for all inorganic nitrogen components during the daytime (Fig. S2)." (L.260-261, p. 9).

○Line 326. I am not convinced the authors' argument here is correct. I would expect, to the first order, HNO3 and NH3 to be driven off the aerosol in stoichiometric ratios and thus the flux divergence for both compounds (and their aerosol counterparts) should be similar in absolute (molar) terms, independent of the deposition rate of the individual compounds. However, Fig. 5 shows normalised (i.e. relative) concentration changes and here the authors are correct with their second explanation: the relative effect on NH4+ is smaller than on NO3- because it partly represents non-volatile sulfates.

Response: Our statements were somewhat confusing. We removed it and added the sentences accordingly: "NH4+ flux over the forest was less influenced than NO3- by gas-to-particle conversion. It is because dry deposition rates of NH3 were much lower than those of HNO3, and so differences in deposition rates between NH3 and NH4+ are much smaller than those between HNO3 and NO3." (L.382-384, p.13).

Technical corrections:

○Title. The word "in" does not read right in my mind. How about "The effect of aerosol dynamics and gas-particle conversion on dry deposition …"

Response: Thank you for your suggestion: I modified the title as "The effect of aerosol dynamics and gas-particle conversion on dry deposition of inorganic reactive nitrogen in a temperate forest".

○Line 88. "fluxes with stomata … and with leaf water surfaces …"

Response: We replaced "over" to "with" as you suggested.

○Eqs. (1), (2), (3), (5). Please make sure the text introduces all symbols used in the equations. Many symbols (e.g. R, a, Gs) are not introduced.

Response: The variables are not introduced as you suggested. We checked the equations and added the explanation for a, R', Gs (with the appropriate reference; Nemitz et al., 2004).

○Eq. (3) would benefit from a reference.

Response: We gave two references for this equation (Nemitz et al., 2000; Sutton et al., 1994).

○Eq. (5). Is the meaning of a here the same as in Eqs. (1) and (2)? If not, please use a different symbol.

Response: The explanation of "a (leaf area density)" was missing; as other reviewer also suggested, we introduced this variable just after Eqs. (1) and (2).

○Eq. (5) and (6) and associated text. It would help here to make any dependence on particle diameter explicit, e.g. by writing Ep(Dp).

Response: As you suggested, we modify both equations (currently Eqs. (7) and (8)) with dependence of particle diameter (Dp) with an explanation.

○Line 130. "… transfer is driven by the difference …"

Response: We modified this sentence as you suggested.

○Line 173. It is not clear to me what the word "latter" refers to. Please rephrase.

Response: The word "latter" was rephrased as "incoming long-wave radiation". (L.199, P.7)

○Line 190. It would be useful to state the total number of layers of the model.

Response: As suggested by other reviewer, we added the information of layer number in Table S1 in supplement.

○Line 214. The acronym "gpc" is not introduced. Presumably it stands for gas-particle conversion? I wonder whether "thermodynamic gas-particle partitioning" would be a better concept to use throughout?

Response: We should introduce the acronym in the manuscript. As you suggested, we define the word gpc as "thermodynamic gas-particle partitioning" (L.XX, p.YY).

○Figure 6. Please clarify in the figure caption the height for which this flux is provided as the flux is height dependent. In addition, it may be illustrative to display the actual exchange with the ecosystem under both model scenarios.

Response: We added the height (30 m) for output of (apparent) mass flux in the caption of new Fig. 8. Furthermore, "The actual deposition flux of each component by forest (ecosystem flux) was also shown for comparisons with apparent flux." (L.42, p.2).

○All figures: horizontal zero lines on all figures would help interpret these more easily. The font size of some labels and legends should be increased for better readability.

Response: We increased font sizes and added zero lines to all figures.

○Line 322. This should read "2004" rather than "2004a" here, I believe.

Response: It was corrected to "2004". Thank you so much.