Refee #2

General comments:

This manuscript presents the results from the HTAP II multi-model study on the impact of hemispheric transport to the atmospheric deposition of S and N. Compared to HTAP I studies HTAP II has different definition of regions with higher spatial resolution which provides more accurate information for coastal regions. The manuscript falls within the scope of the journal and is suitable for publication in ACP after a number of corrections that will improve its clarity and are listed below:

Response: We would like to thank the reviewer for the suggestions to improve the manuscript. Following are the point-by-point responses to the comments.

Specific comments:

1- Explain why the specific regions have been chosen for the study.

Response: According to the design of the HTAP II perturbation experiment as shown in Figure 2 in (Galmarini et al., 2017), the six regions used in this study (NA, EU, EA, SA, RU and ME) are of the highest priority in the model simulations. The participating models are required to submit the model results under perturbation experiments of these regions. However, we find some other regions like North Africa and Southeast Asia might also be largely affected by long range transport. And we suggest to include more regions in future simulations in the last paragraph of the conclusion section.

2- Title: Source contributions TO sulfur and nitrogen deposition.

Response: We have changed it.

3- line 34- abstract (but also elsewhere in the text)- west East Asia does not read nicely  
Response: We use abbreviation EA for East Asia. The “west East Asia” is changed to “west EA”.

4- Line 42: 40% and 23-45% do not add up to 100%, where the remaining at least 15% comes from?

Response: The remaining 15% comes from other sources. We explain the possible reason in the manuscript:

“It could come from the emission reduction in rest of world, especially nearby regions such as from Central Asia and North Africa to EU and ME. It could also come from the joint effects of emission control by multiple source regions, which possibly change the oxidant chemistry, atmospheric mixing and lifetimes of reactive pollutants. However, the model simulations do not allow to separate these two contributions in this study.”

5- Line 68: remove ‘to’ after ‘3-5 ppb’

Response: We have changed it.

6- Line 77: ‘significant’; can you derive a number or better a range of numbers for some pollutants from the publications? to quantify the ‘significant’ line 87: used

Response: We collect the range of impact from publications and rewrite the sentences.

“Recent studies have reported an increasing trend in the hemispheric transport of air pollution from Asia to NA from mid-1980s to late-2000s. The Asian plume has contributed ~10 ppbv (30%) to the O3 concentration over western NA from mid-1980s to mid-2000s (Jaffe et al., 2003; Parrish et al., 2004), with an annal increase of 0.34-0.50 ppbv O3 (Parrish et al., 2009). More recent study showed the contribution is about 5-7 ppbv O3 in 2006 with an annual increase rate of 1-2 ppb O3 since 2000 (Zhang et al., 2008). The trend well agreed with the rapid growth of Asian emission (Richter et al., 2005;Lu et al., 2010;Zhang et al., 2007;van der A et al., 2006;van der A et al., 2008)”

7- Line 160: please clarify whether for the NH4+ wet deposition at stations you compared the modeled NHx deposition with the NH4+ wet deposition measurements, or the NH4+ modeled only. The NH4+ wet deposition measurements are representative of the sum of NH4+ and NH3 deposition.

Response: We have added the following sentences to clarify.

“Modeled gas phase SO2 wet deposition and aerosol SO42- wet deposition are evaluated with observed SO42- wet deposition.”

“Modeled gas phase HNO3 wet deposition and aerosol NO3- wet deposition are compared with observed NO3- wet deposition.”

“Modeled gas phase NH3 wet deposition and aerosol NH4+ wet deposition are compared with observed NH4+ deposition.”

8- Line 164: The most accurate comparison would have been that between atmospheric observations of gases and aerosols with the modeled concentration levels.

Response: We have compared both air concentration and dry deposition velocity between CASTNET and multi-model results. The results are shown in our previous paper “Multi-model study of HTAP II on sulfur and nitrogen deposition”. Following are the related paragraphs in that paper for reference.

“Since the CASTNET dry deposition is not actually measured but instead a calculation of measured concentration of species and modelled dry deposition velocities, it is necessary to investigate which factor of these two contributes to the model bias. We compare the modelled air pollutant concentrations with CASENET measurements as shown in Table S4-S8. The MMM overestimates the SO2, SO42-, HNO3, NO3- and NH4+ concentrations by 394%, 40%, 217%, 135% and 173%, respectively. It should be noted that the CASTNET sites are generally located in rural regions that are away from emission sources (Sickles and Shadwick, 2008), thus the measured concentrations of air pollutants are relatively low compared with those of urban sites. While the resolutions of the HTAP II models range from 0.5° to 3°, and are not fine enough to reproduce the characteristic of some rural sites. The models with finer resolutions except CHASER\_t106 model (i.e. EMEP\_rv48 (0.5° × 0.5°) and SPRINTARS (1.1° × 1.1°)) generally perform better than the others, while models with coarse resolutions (i.e. CHASER\_re1 (2.8° × 2.8°) and OsloCTM3.v2 (2.8° × 2.8°)) are generally not performing well for all species. This could explain the overestimation of air pollutant concentrations at the CASTNET sites.

In order to check the differences of modelled dry deposition velocity between CASNET and HTAP II models, we adopt the general approach for calculating dry deposition velocity from Wesely, (1989). Vd = -Fc /Ca (7), where Vd is the deposition velocity, Fc is the dry deposition flux and Ca is the concentration of species. The negative mark indicates the direction of the dry deposition velocity. This scheme has been widely adopted in global models (Wesely and Hicks, 2000) with modifications. We compare the calculated dry deposition velocity of models and CASTNET (Table S9-S13). The mean bias of dry deposition velocities for MMM are -8%, 0.3%, 7%, 19% and 2% for SO2, SO42-, HNO3, NO3- and NH4+, respectively, which are much lower than those of air pollutants. The model bias for dry deposition at the CASTNET sites mainly comes from the model over prediction of air pollutant concentration.”

To make it clear, we add short description in this manuscript as follows:

“The CASTNET data is calculated with observed aerosol concentration and modeled dry deposition velocity, therefore it has high uncertainty in data quality. Evaluation shows that the modeled dry deposition is generally higher than the CASTNET inferential data by a factor of 1-2. This is a common feature of many global and regional models (WMO, 2017). According to the analysis, the model bias for dry deposition at the CASTNET sites mainly comes from the model over-prediction of air pollutant concentration. The CASTNET sites are generally located in remote regions with relatively lower air pollutant concentrations than urban regions, but the models fail to represent this characteristic with coarse spatial resolution (Tan et al., 2018).”

9- Lines 179-180: mass balance requirements. You need to provide more information specific to this study and mention here what tolerance you have for the mass balance. Is it for instance no tolerance at all? 1 per mile? or higher? This means that you also need to define what means ‘almost identical’ (line 184).

Response: We added the criteria in the manuscript.

“We compare the global total amounts of changes of deposition (∆ Depo) with changes of emissions (∆ Emis) for all perturbation cases (Table S1). Models are excluded if their global ∆ Depo values fall outside the range of ±20% of their global ∆ Emis.”

10- Lines 185-186: Can you explain why the mass balance requirements are not fulfilled for one region while they are fulfilled for the others? What is the particularity of EA?

Response: Sorry for the confusion. The change of deposition under EA case is missing because no model has submitted the wet deposition of NH4+ under the EA perturbation case, although they have submitted the data under base case. We have rephrased the sentence in the manuscript.

“The ∆ Depo of NHx deposition under EA case is not available due to lack of model results for the wet deposition of NH4+ under 20% emission perturbation in EA.”

11- Line 211: please provide a comment on the sources that lead to injections of S emissions higher than NOx emissions in the models. (relevant discussion line 315 – please provide some numbers)

Response: The injection heights of S and NOx emission are different in the emission inventory due to their emission sources. The following figure shows the contribution of emission sectors to air pollutants in the emission inventory of HTAP II (Janssens-Maenhout et al., 2015). SO2 mainly comes from power plants and industry (~80%). Both of these two sectors have high outlets. For NOx, the contribution of these two sectors are much lower (~40%). About 40% of NOx is from transport section, which emits pollutant near the earth surface. Thus, we conclude that the S emissions are emitted from higher altitude than NOx emission. However, we notice that some global models (such as CAM-Chem and C-IFS\_v2 models) treat power plant emissions at first layer. In that case, the injection height won’t affect the results, but the generally longer lifetime of S emission could affect the results.

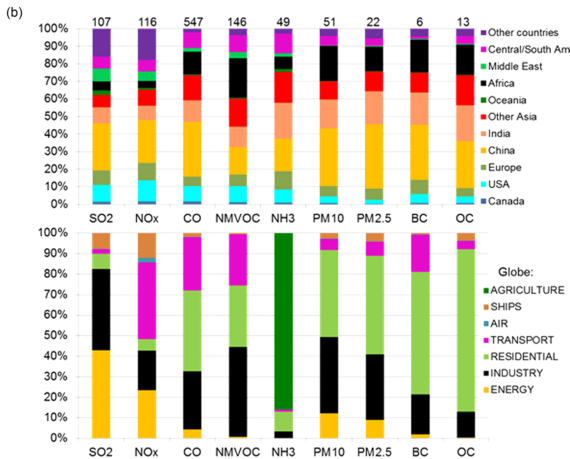


Figure from (Janssens-Maenhout et al., 2015) Global sector-specific anthropogenic emissions of gaseous pollutants and particulate matter components for the year 2010. Global absolute emissions are reported on top of each bar in Tg species per year. Large scale open-biomass burning is not included in the analysis.

12- Line 225: from the atmosphere

Response: We have changed it.

13- Line 226 ‘summer’ do you mean winter?

Response: We notice that summer or winter is not the rainfall season for all regions. We deleted it in the manuscript.

14- Lines 227-228: indicate the region to which you refer?

Response: We rewrite this sentence for clarification.

“In addition, the strong westerly winds in winter and spring favor the hemispheric transport for regions in mid-latitudes of the North Hemisphere. While the rapid vertical convection in summer slows down the zonal transport of air flows and accelerates the local removal process.”

15- Line 231: IN our study

Response: We have changed it.

16- Lines 236-237: please rephrase in this sentence it is not clear which is what.

Response: We have rephrased the sentence.

“HTAP I study by (Sanderson et al., 2008) developed a SR relationship for NOy deposition among NA, EU, SA and EA. Their results showed that about 12-24% of the emitted NOx is deposited out of source regions. This study of HTAP II finds a higher percentage of export (26-34%).”

17- Line 239: comparable:

Response: We have changed it.

18- Lines 267-273: please comment on which circulations patterns contribute to these impacts.

Response: We find the circulation pattern is strongly related to the seasonality of long-range transport. So we described the circulation with seasonality in another paragraph in line 340-356.

“The deposition change via transport between neighboring regions is found throughout the whole year and is slightly stronger in winter, such as between EU and RU (~30°E) (Fig. 4(b) and (f)) and from EA to the North Pacific Ocean (~130°E) (Fig. 4(d)). This is consistent with the seasonality we found for the export of emission by source regions in section 3.1. In addition, most source regions reduce more S and NOx emissions in winter than the other seasons (Table S3), thus more emissions are exported abroad in winter. On the contrary, most of the deposition change by transport over long distance occurs in spring and fall, especially for the hemispheric transport from NA to EU, from EU to EA and from EA to NA. The seasonality of long-range transport for NA, EU and EA well fits the characteristic of westerlies, which is the prevailing winds in the mid-latitude of the North Hemisphere. This agrees with the seasonality of the transpacific, transatlantic and trans-Eurasia flows of air pollutants that spring is the most efficient season for long-range transport for mid-latitudes. (Holzer et al., 2005;Liu et al., 2005;Liang et al., 2004;Brown-Steiner and Hess, 2011;Li et al., 2014;Auvray and Bey, 2005;Wild et al., 2004;Liu et al., 2003). Although the westerlies is also strong in winter, the long distance transport of emissions is low, because the formation of secondary species like PAN is suppressed by slow oxidation in cold environment (Berntsen et al., 1999;Deolal et al., 2013;Moxim et al., 1996), which plays an important role as a reservoir for NOx in the long-range transport of air plumes (Lin et al., 2010;Hudman et al., 2004).”

19- Line 275: ‘impact similar to that of S emission’ does this mean similar lifetime of SOx and NOx ? if yes is this consistent with discussion in lines 211 and 315?

Response: No, S has longer lifetime than NOx. We rewrite the sentence.

“The overall impact is qualitatively similar to that of S emission in the spatial pattern, with some differences in the values.”

20- Lines 278-288: Could you comment on potential differences in aerosol pH that impact on the NOy partitioning to the aerosol phase?

Response: This is an interesting question. The pH value affects the dissolution of gas-phase HNO3. The gas-phase HNO3 is produced by daytime reaction Eq. (1), and is very soluble in water (Eq. (2)) and dissociates to aerosol phase NO3- (Eq. (3)). Under the ideal solution, the higher the pH value, the more gas-phase HNO3 can be dissolved.

NO2 + OH → HNO3 (gas) (1)

HNO3 (g) ⇌ HNO3 (aq) (2)

HNO3 (aq) ⇌ NO3- + H+  (3)

In this case, the perturbation in SO2 emission decreases the concentration of acid SO42- in aerosol, which increases the aerosol pH value. If ignore the effects on the dissolution of NH3, this condition will increase the fraction of NOy partitioned to aerosol phase. In addition, study by (Bian et al., 2017) found that the NH3 wet deposition is very sensitive to the pH value in the cloud, which as a result will largely affect the NH3-NH4+-NO3- equilibrium. The models without pH adjustment can provide very different results of NO3- aerosol with models that with the adjustment.

21- Lines 324-327: briefly describe the reported seasonality.

Response: We have changed in the manuscript.

“This agrees with the seasonality of the transpacific, transatlantic and trans-Eurasia flows of air pollutants that spring is the most efficient season for long-range transport for mid-latitudes.”

22- Line 328: replace ‘due to that’ by ‘because’

Response: We have changed it.

23- Lines 361-364: can you discuss this based on atmospheric circulation?

Response: We have added the following description for circulation.

“Except large scale circulation like prevailing westerlies, the coastal regions are featured with complex small scale circulations. For instance, the low-level jet (zonal winds with high speed) contributes to the rainfall in coastal regions in Asia (Xavier et al., 2018). The orographic effects enhance the precipitation over coastal mountain regions such as west coast of NA, EU and southeast coast of RU (James and Houze, 2005).”

24- Line 381: ‘other reasons’ & line 388 ‘joint effects’ : can you spell out potential reasons? Could this be change in oxidant chemistry and lifetimes for instance?

Response: we explain the “other reason” in following sentence:

“It indicates the deposition change due to other reasons than the total effects of separate emission reduction in the 6 regions. It could come from the emission reduction in rest of world, especially nearby regions such as from Central Asia and North Africa to EU and ME. It could also come from the joint effects of emission control by multiple source regions, which possibly change the oxidant chemistry, atmospheric mixing and lifetimes of reactive pollutants. However, the model simulations do not allow to separate these two contributions in this study.”

25- Line 397: separately

Response: We have changed it.

26- Lines 402-404 are very hard to read, please separate and rephrase this sentence for clarity

Response: we have rephrased the sentence.

“In terms of S deposition (Fig. 7(a)), the ranges of modelled ∆ Depo by multiple models, defined as (modelled maximum value – modelled minimum value), are (0.06-0.23) Tg(N) yr-1 and (0.01-0.22) Tg(N) yr-1 for SO2 dry and wet deposition, and (0.01-0.03) Tg(N) yr-1 and (0.009-0.17) Tg(N) yr-1 for SO42- dry and wet deposition, respectively.”

We also change the corresponding sentences for NOy and NHx deposition.

“In terms of NOy deposition (Fig. 5(b)), the differences among models range (0.003-0.07) Tg(N) yr-1 for NO2 dry deposition, and (0.07-0.55) Tg(N) yr-1 and (0.03-0.75) Tg(N) yr-1 for NO3- dry and wet deposition, respectively.”

“In terms of NHx deposition (Fig. 5(c)), the differences among models range (0.04-0.09) Tg(N) yr-1 for NH3 dry deposition, and (0.008-0.15) Tg(N) yr-1 and (0.002-0.11) Tg(N) yr-1 for NH4+ dry and wet deposition, respectively.”

27- Lines 402-412: I think the units should be Tg(S)/yr or Tg(N)/yr.

Response: We have changed it.

28- Lines 427-429: you provide percentages for export of emissions from Europe to Russia and hen from Russia to Europe; similar for EA to RU and RU to EA, why you do not provide the net effect? Please rephrase for clarity.

Response: The export fraction are related to the total emission amount and seasonal variation of the source regions, thus we prefer to describe the fraction separately for each region. In order to make it clear, we rewrite the sentence:

“The most significant exports of emissions are: 1) transport between EU and RU. 10-14% of EU’s emission is transported to RU and 7-12% of RU’s emission is transport to EU. 2) transport between EA and RU. 5% of EA’s emission is transported to RU and 4-5% of RU’s emission is transported to EA. 3) transport from SA to EA (4-9%).”

29- Lines 429-430: remove ‘to abroad’ since it is export.

Response: We have changed it.

30- Line 447: do you mean that ‘coastal regions receive upmost half of their deposition though hemispheric transport from foreign regions’ or that ‘half of the transported by LRT amount from foreign regions is deposited over the coastal regions’ ?

Response: The later one is the appropriate understanding. We rephrase the sentence in the manuscript.

“For some regions, upmost half of the hemispheric transport from foreign regions is deposited over their coastal regions.”

31- Figure captions:

Response: We have changed it.

32- Line 745: correct MD to ME

Response: We have changed it.

33- Figure 1: why this figure does not show the upper boundaries of regions 3,4,14 and the Arctic?

Response: We re-do the figure for clarity. The upper boundaries of regions 3,4 and 14 are region 16-Arctic.

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34- Line 751: add per 0.1x0.1 deg grid box

Response: We have changed it. We also change the unit in the caption of figure 3.

35- Line 4: According to eq 1 the negative values should indicate increase in deposition with decline in emissions

Response: Yes, thus table 1 are all possible values, which means decrease in emission lead to decrease in deposition.

Reference:

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