"Impact of future land cover changes on HNO_3 and O_3 surface dry deposition"

by T. Verbeke et al.

Response to the reviewers – 18th November 2015

We wish to express our sincere gratitude to the two Referees for their careful reading and their constructive and very valuable feedbacks. This definitely helped us to clarify and strengthen the presentation of our work and to improve our manuscript. We took into account thoroughly most of the corrections and suggestions and hope that this new version of the manuscript will meet, if not all, most of the Referees' expectations.

Anonymous Referee #1

SUMMARY

This paper describes a modelling study of ozone and nitric acid dry deposition in a state-of-science global chemistry-climate model. The main focus is on assessing the impact of land cover changes (LCC) on O_3 and HNO_3 dry deposition between presentday (2000s) and the future (2050s). To cover a wide range of potential changes three of the four CMIP5 RCP scenarios are evaluated. The impact of LCC and climate is assessed separately.

GENERAL COMMENTS

My expectations for this paper were high, maybe a bit too high, because the process of ozone dry deposition at the surface is crucial for the understanding of atmospheric composition and the consequences for climate and air quality. Combining that with nitric acid dry deposition the paper promised to deliver a really interesting study.

To a fairly substantial degree, my expectations have been fulfilled. Attribution of future changes in O_3 and HNO_3 dry deposition to either LCC or climate change have been assessed and relative magnitude of both effects have been discussed. This makes this paper a valuable and very relevant addition to science.

My main reservation then is with the execution. To me the paper feels somewhat unfinished and unpolished. The paper lacks structure and depth. Too many findings are just discussed "in passing" with rather little attention to the details, it seems to me at least.

Here is what I'd like this paper to include:

+ clear division between annual mean impacts and seasonal variation + more detailed discussion of the consequences for atmospheric composition, specifically, in terms of atmospheric oxidative capacity (OH concentration), air quality and possibly exceedances (surface ozone), and potential health and ecosystem impacts (not necessarily in quantitative terms, a qualitative discussion only would already be nice) + similarly for HNO_3 in terms of acid deposition damage and N-fertilisation potentials. + analysis of the relative importance between aerodynamic and surface resistance terms to dry deposition (conceding that quasi-laminar resistance may not play a decisive role, but I do not know for sure), potentially on a per PFT basis.

Looking at this list, I appreciate that I may be asking too much, as I have already mentioned above. Let me go through the individual sections to make it a bit clearer (hopefully) what I mean.

The introductions are sound and adequate, nothing to complain there, and so are the sections on model description and description of the dry deposition scheme in LMDz-INCA. The description of the LCC changes between PD and FU and the modelling strategy also have their merits.

When it comes to discussing the findings, though, the paper, to some degree, comes undone. PD ozone and HNO3 deposition is discussed first but a bit superficially for my taste. I'd like to see the following subsections: 3.1.1 spatial patterns of O3 and HNO3 dry deposition under PD conditions based on annual means using the regions defined in the paper. 3.1.2 seasonal variation in the deposition fluxes for O3 and HNO3 (using the proper seasonal means DJF, JJA, etc., and not just monthly means for January and July, to give an example). 3.1.3 a brief discussion of the relative importance of the individual resistance terms, maybe with respect to location, vegetation composition (grassland versus forest, for instance) and seasonal variation would be extremely nice.

The next section discusses changes between PD and FU scenarios. A similar structure seems appropriate. I could imagine the following subsections: 3.2.1 changes in the annual mean deposition fluxes with respect to specific characteristics which manifest due to the three CMIP5 RCP scenarios. 3.2.2 the impact of seasonal variability 3.2.3 if apparent at all, a brief discussion of the changes in the leading terms between PD and FU, i.e., are the same resistance terms dominant or not.

Section 3.3 focuses on impacts on atmospheric composition. However, this section is rather brief and a little bit unfocused. Possible questions to address here are impacts on the a) ozone budget, b) the OH concentration and c) surface ozone concentration with a view to air quality and health/ecosystem damages. I am thinking of no more than four or five paragraphs in total for this section.

Section 3.4 is simply too short for me taste. Here, an attribution to LCC and climate change is attempted but not really discussed at any length. Just use more of the information available from the experiments.

The "Discussion and conclusions" section is quite reasonable. It could also be improved, however, by discussing in more detail what the newly added sections have revealed, i.e., the contrast between winter and summer seasons, its change with climate change and some conclusions on what that could mean to future air quality and possibly human health and ecosystem functioning.

Once completed, these changes can also be percolated to the abstract making it stronger and more captivating, too. Anyway, these are just my thoughts.

→ *We tried to expand the discussion of the results based on all this remark. However the aim of the paper, which is now more explained, is to isolate specifically one effect (LCC) on one process (dry deposition) and to see if it is, or not, necessary to keep attention of this when studying the atmospheric chemistry in the future. We show that the answer to this question is positive but, as discussed by Hardacre et al 2015, it first requires to go into details in the hypotheses done by in model to consider the land cover repartition. Beyond that point, we don't want to go too deep in the results on atmospheric chemistry because some of the impacts (in particular on OH) are highly model dependant and because we think that without considering all the potential vegetation/atmospheric chemistry (now shown in Figure 1), it does not really make sense to consider all the chain of consequences. The conclusion of the paper is thus rather a methodological recommendation than the quantification of an effect. Regarding the* *seasonal variability, we show in the supplementary material the sesonnal mean rather than monthly mean. However, the way the sesonality of vegetation is considered in the model does not reflect for example the LAI changes thrghout the year. For that reason, a delayed analysis of all the processes and their changes by season could be confusing for the reader.*

I concede that hindsight is always perfect and that it is far more easy to criticize than to create. Hence, I suggest the paper to be accepted with minor changes only which hopefully will reflect some of the suggestions that I have made in the general comments.

What follows are some specific comments which mainly pertain to typos and such like.

SPECIFIC COMMENTS

p.18461/l.25: the symbol for the element sulphur, “S”, is a non-italic.
→ *Changed*

same/l.26-27: the sentence need editing.
→ *Modified*

p.18642/l.22: should read “beginning of the industrial era”
→ *Changed*

p.18463/l.5: should read “oxidative capacity” or “oxidizing capacity”
→ *Changed*

same/l.8: better: “...as given in the three RCP scenarios. . .”
→ *Changed*

same/l.20: citation “Lamarque et al., 2010” seems to be missing from the reference list
→ *Added*

p.18465/l.6: “exposed” - better: “highlighted” or “depicted” or “shown”
→ *Changed*

same/same: better: “...and Europe as presented in. . .”
→ *Changed*

same/l.27: citation “Loveland et al., 2000” seems to be missing from the reference list
→ *Added*

same/l.28: citation “Dufresne et al., 2013” seems to be missing from the reference list
→ *Added*

p.18467/l.10: reference “See et al., 2011” seems to be missing from the reference list
→ *Added*

same/l.23: changes in temperature, strictly speaking, are measured in Kelvin

→ We kept the temperature change mentioned in °C, as also used in international reports such as the IPCC ones.

same: should read “mean surface temperature change is 0.93K between. . .”
→ Same as above

p.18468/l.8: should read “sub saharan Africa”
→ Changed

same/l.26: reference “Walcek et al., 1986” seems to be missing from the reference list
→ Added

p.18469/l.10-11: “For HNO3. . .”; sentence needs revision
→ rephrased

same/l.19: should read “in terms of”
→ Changed

p.18471/l. 7: should read “the LCC effects”
→ Changed

same/l.12: better “The RCP8.5 scenario leads to...”
→ Changed

same/l.12-13: better “due to the reduction in the deposition rate”
→ Changed

same/l.22: better “...an increase of the HNO3 deposition flux...”
→ Changed

same/l.24: better “It thus leads to a reduction in the HNO3 concentration by 0.2 ppb(v)...”
→ Changed

p.18472/l.4: should read “To this purpose,...”
→ Changed

same/l.5: better “0.93K”
→ We kept the temperature change mentioned in °C, as also used in international reports such as the IPCC ones.

same/l.6: better “...temperature increase projected in the RCP scenarios...”
→ Changed

same/l.7: should read “...this climate change on the deposition rate. . .”
→ Changed

same/l.10: should read “The climate effect on the deposition rate. . .”
→ Changed

same/l.12: better “solar irradiance”
→ Changed

same/l.23: better “Discussion and conclusion(s)”
→ Changed

same/l.21: should read “...to assess the impact of changes. . .”
→ Changed

p.18473/l.16: should read “..., for both of the...”
→ Changed

same/l.23: better “0.93K”
→ We kept the temperature change mentioned in °C, as also used in international reports such as the IPCC ones.

same/same: should read “..., we calculate that. . .”
→ Changed

p.18474/l.3: should read “..., and lack of the representation of. . .”
→ Changed

same/l.7: better “...proper input parameters for dry deposition schemes, . . .”
→ Changed

References:

p.18475/l.9: reference “Hurtt et al., 2010” does not seem to be cited in the text
→ Changed

Table 2 on p.18478: not all changes higher than 1.5% are highlighted in the table
→ Modified to highlight all the occurrences

Anonymous Referee #2

General Impression

The manuscript by Verbeke et al assesses the impact on land cover change on the deposition velocity and, by association, deposition and air concentration of HNO3 and O3 and tries to compare the impact with the isolated impact of climate change. The subject of interest and worth of publication, but the current manuscript lacks clarification of what was actually done, a discussion of the uncertainties and limitations of the approach as well as a discussion of the results in the context of other studies. It therefore requires significant modifications before it can be accepted for publication in ACP. I am very disappointed that most of my technical comments provided at the access review stage for ACPD have not been taken on board, including some clear spelling mistakes which I now have to repeat here.

Major comments

1. As with any Chemistry Transport Models and even more so, Earth System Models, there are hidden interactions within the model that complicate the interpretation of the results and much depends on the exact implementation. Looking at the impact of one change (LUC or climate) in isolation is difficult both technically (because of the links within the modelling system) as well as philosophically (because in reality changes do not occur in isolation). Therefore, for the reader to be able to understand the implications of the work, it is essential that the authors provide as much detail as possible on what which part were kept constant between runs and what was changed. This should be added to Section 2. At present much of the detail is obscure or is clarified very late in the manuscript:

> a. The authors should provide some more detail on the Wesely parameterisation and clarify further to what extent their implementation matches that of Wesely. For example, what parameterisation is used for Rb, the values of which differ between parameterisations in the literature, especially for aerodynamically rough surfaces (i.e. forests).
>
> → *Ra and Rb in LMDzINCA are calculated based on Walcek et al. (1986). This information was indeed missing and is now added in the text. Complementary information regarding the deposition schema in LMDzINCA is now also included in the section 2.1.*
>
> b. For example, no information is given on the calculation of the stomatal conductance and the authors should confirm in the manuscript that this follows Wesely as it is often generated independently in CTMs. If it does follow Wesely (as implied only by the very last paragraph of the discussion section), it is not increased by CO2-related increases in gross primary productivity or lowered by the ability of plants to reduce water loss whilst maintaining photosynthesis under higher CO2 concentrations. The feedback of changes in O3 itself on plant growth and stomatal conductance (Sitch et al., 2007) is then also not captured.
>
> → *In LMDzINCA, the stomatal resistance is determined following the Wesely et al. (1989) approach and potential important interactions with the environmental conditions are indeed not taken into account. This is now stated in the description of the deposition scheme, section 2.1.*
>
> c. Similarly, it is unclear whether the model calculates a single u* and heat flux for each grid cell or whether this is calculated for each land cover class individually. If the former, the implications need to be discussed. Landcover change changes the roughness and heat fluxes at the landscape scale, which in reality feeds back on the meteorology. However, the authors seem to drive the LUC scenarios nudged with the same meteorology, which generates an inconsistency. Would it not be more appropriate to do free 10-year runs GCM runs similar to the climate change scenario runs, but keeping atmospheric composition and sea surface temperature and also ice cover (which is not mentioned, but is presumably changed between the GCM runs?) constant. For these runs the LUC would have the ability to feedback on climate.
>
> → *Following the approach by Walcek et al. (1986) the aerodynamic and laminar deposition resistances Ra and Rb are calculated for each land-type, mainly through the use of a roughness length that is prescribed for each land-type and season categories. We totally agree that in reality, a change in land-type therefore has a potential impact, through modification on the roughness and heat flux, on meteorology. We appreciate that this is a limitation of our study but our objective here is only to consider the one-way change of deposition in relation with land-*

cover change, rather than to provide a comprehensive picture of every feedbacks involved. This is now specified and discussed in the manuscript.

d. Agricultural emissions appear not to have been changed between runs. In reality, LUC induces a change in both natural and anthropogenic emissions. Therefore, it is more meaningful to compare changes in Vd than in concentration and deposition.

e. Whilst BVOC emissions appear to be kept constant between runs, presumably, the deposition of other compounds, that chemically interact with HNO3 (e.g. NH3) and O3 (e.g. NO2 and VOCs) also change with landcover and climate. Thus, the change in concentration is no longer merely affected by the deposition velocity of the compounds themselves.

→ *Emissions from every source, including agricultural, biogenic and anthropogenic ones, are indeed kept constant between runs, which is inconsistent with the different changes in surface occupancy considered in our study. Rather than to build a real case scenario considering changes of all possible parameters involved, our objective is to perform a first-step investigation, analyzing the potential impact of changes in land-use on atmospheric chemistry, only through the mechanism of changes in surface dry deposition. It is completely true that in a real world, such changes in land surface type and area would go together with changes in climate, emissions, and therefore atmospheric chemical mechanisms as well. More effects would therefore need to be considered to get a better picture of the actual overall change. We agree totally that other important impacts would play a key role and would lead to a different response but our aim is to isolate the change in dry deposition velocity, deposition, and concentrations as well, related to land-cover change only, which is still poorly understood and quantified and therefore needs to be addressed specifically. These important elements are now added in the manuscript (Introduction, modeling set up and conclusion/discussion sections).*

2. Comparison with measurement data, limitations of the Wesely approach.

a. The paper lacks any sort of assessment of the modelled Vd with measurements (or reference to another paper that performs this assessment) and it is therefore difficult to assess how the parameterisation (and its implementation into this particular CTM) performs under current conditions and whether the predicted changes are therefore robust. The Wesely approach is now 15 years old and does not reflect the measurement evidence of the past decades, with several studies indicating, e.g., that deposition rates to wet cuticles are larger than to dry surfaces, although the process understanding is still uncertain. The Wesely approach greatly relies on static look-up table derived from measurements under current conditions which may change in the future. For example, it does not include a mechanistic understanding of the effects of leaf water chemistry on non-stomatal pathways, which may be altered by climate and composition change. None of these uncertainties and limitations of the study are discussed in the present manuscript.

→ *Regarding dry deposition, the lack of measurements, and the technical challenge to measure the relevant variables in field or laboratory conditions, make it difficult to evaluate models properly. Such an evaluation regarding the LMDzINCA ability to quantify dry deposition of atmospheric chemical species has therefore not yet been performed in details. However, several papers already published demonstrate the performance of the model to adequately represent key chemical processes, including sources and sinks, and dry deposition especially (Hauglustaine et al., 2004; Folberth et al., 2006). Several elements on this topic are now added in the manuscript,* *and especially in the scheme description section 2.1, together with an extended discussion in section 4 on the limitations of the model with regard to recent key findings published in the literature. To allow an evaluation of LMDzINCA performance in term of dry deposition estimation in comparison with other models, we also included a new section in the Supplementary Material "A. Dry deposition evaluation". The added Figure S1 illustrates the ozone flux calculated by LMDzINCA for several locations, and comparison with other model estimates, based on the work recently published by Hardacre et al. (2015), as suggested by the referees.*

b. It seems counterintuitive why crops should provide a more efficient sink to ozone than coniferous forest, especially in winter (P18468 and first figure in Supplement), because the "LAI" of bare soil is much smaller than that of forest. Also, at present it is not clear whether the figure in the supplement reflects the global average (i.e. mixes winter and summer values) as no caption is provided.

→ *Following the approach developed by Wesely (1989), the leaf area index is not directly taken into account in the calculation of resistances, which are therefore strongly determined by land-cover types, with reference values being prescribed depending on vegetation type. The relation between plant surface and resistances is represented in LMDzINCA by the use of a seasonal index, as described in section 2.1. This is indeed a limitation of such deposition schemes, and other approaches have been published in the literature, such as the parameterization proposed by Zhang et al. (2003). We add this important point, which was not addressed so far in details, in the manuscript. Captions are now added to the figures of the supplementary material.*

3. I would have expected the authors to discuss findings in much more detail in relation to the published literature. How large are the expected concentration changes due to LUC compared with other effects, such as changes in emissions and chemistry? Some key references such as Hardacre et al. (2015) are not discussed.

→ *Despite a few studies have been carried out recently on the dry deposition changes or land cover change effects on chemistry in the future, none of them try specifically to assess the land cover change impact on dry deposition. We added a figure (Figure 1) showing the main interactions between vegetation and atmospheric chemistry in order to indicate more precisely what we do aim in this study and how the effect we are looking for takes place in this general framework. We also added the reference to several recent studies. Furthermore, we added a comparison with Hardacre et al. 2015 in the supplementary material.*

Minor Scientific Comments

1. I suggest the authors avoid the use of the term "deposition rate" throughout the manuscript (text & figures), because this is used ambiguously in the literature, sometimes referring to the deposition flux and sometimes to the deposition velocity. I suggest you use "deposition velocity" throughout.

→ *This is now corrected throughout all the manuscript*

2. In the abstract, please point out that the same meteorology was used for the LUC scenarios.

→ *This information is now added in the abstract*

3. Vd and Ra are both height dependent, which should be indicate as Vd(z) in Eq. (1). Please state throughout the height to which the values of Vd refer to throughout the manuscript and in the captions to the appropriate figures.

→ Dry deposition at the surface is investigated throughout the manuscript. This information is now clarified in the text, including the abstract and the supplementary material, and the figure captions.

4. Is it really the oxidation capacity that is treated by Wesely? I thought it was more generally the reactivity.

→ As clarified in the text "R_c is based on the temperature dependent Henry's Law effective coefficient and reactivity factor for the oxidation of biological substances (Folberth et al., 2006)"

5. One of the most important factors affecting deposition rates is turbulence (P18462, L10), which also governs Ra (P18464, L3) and, together with surface roughness also affects Rb (L4). By contrast, Rc is probably more affected by LAI and canopy structure than by roughness length (L7).

→ For clarification and for a better understanding of important parameters, this point has been added in the introduction and in the section 2.1 (dry deposition description).

6. P18471, L4. As mentioned above, this includes the deposition of other compounds that chemically react with O3 and HNO3. This also needs to mention in relation to P18471, L25.

→ It's true. We think that O3 and HNO3 changes are here probably mainly driven by changes in their own dry deposition velocities. However it can not be restricted to that. We thus modified the sentence.

Technical comments (in addition to those already raised by Reviewer 1):

Please sort out the (missing) use of super- and subscripts throughout the document (incl. Supplementary Information and figures), which is very sloppy throughout and distracts from the content. Subscripts are used in the equations, but not in the text. Also, I suggest you write VdHNO3 as Vd(HNO3) or $Vd,HNO3$, ideally even stating the height it refers to.

→ All these points have been checked and modified accordingly in the manuscript and the supplementary information.

Please number figures in Supplement and add captions.
→ Figures number and captions added

Figure 4 and 2nd figure in SM: the units of the deposition flux are still incorrect by many orders of magnitude, possibly the s-1 should read a-1? Please check and correct. Also, usually, these are stated as m-2 (if averages) or maybe ha-1 if annual totals.

→ We thank the reviewer for noting it. The bug in the computation was fixed and all the units are now homogeneously expressed.

Symbols VdO3 and VdHNO3 are not defined in the abstract.
→ Names added in the abstract

P18461, L7: Better English: “… should therefore be considered …”
→ *Changed*

P18461, L10: Plays a key role in what?
→ *Changed*

P18461, L19: Add a reference for Wesely’s description already here.
→ *Reference added*

Table 2. In the header ‘ozon’ should read ‘ozone’.
→ *The spelling was correct in the original manuscript submitted. We will check more carefully in the next version before online publication.*

P18461, L26. Correct spelling of ‘highly’.
→ *Changed*

P18462, L6. Should read “at the regional …”
→ *Changed*

P18462, L14. Should read “in a very simplistic way”
→ *Changed*

P18462, L15. Improve English; two occurrences of “usually” in the same sentence.
→ *Changed*

p18465, L17. Should read "in more detail"
→ *Changed*

p18466, L23. Better: “a strong increase in the cover of all forest categories”
→ *Changed*

p18466, L27. Should read “America loses”
→ *Changed*

p18467, L4. Better “dry deposition without any climate change”
→ *Changed*

p18466, L7. Specify that 2007 meteorology is used.
→ *As the particular meteorology year doesn’t have any impact on the vegetation distribution, we didn’t make this exact modification, but specified that the 2007 vegetation distribution is considered.*

p18466, L23. Should read “temperature change is”
→ *Changed*

p18467, L10. Better “lower annual VdO3”
→ *Changed*

P18469, L18. Should read “by land cover change”

→ Changed

P18469, L27. Should read “decreasing by up to 0.2” and similar instances elsewhere in the manuscript.

→ Changed

P18471, L15. Better “in the east” and “in the west”

→ Changed

P18472, L21. Should read “assess the impact”

→ Changed

P18473, L13. There should be a comma after “Eurasia”

→ Changed

P18474, L11. Should be “show” instead of “shows”

→ Changed

P18474, L14. Should read “The next generation”

→ Changed

References

Hardacre, C., Wild, O., and Emberson, L.: An evaluation of ozone dry deposition in global scale chemistry climate models, Atmos. Chem. Phys., 15, 6419-6436, doi:10.5194/acp-15-6419- 2015, 2015.

Sitch, S., P. M. Cox, et al.: Indirect radiative forcing of climate change through ozone effects on the land-carbon sink. Nature 448(7155): 791-794, 2007.

Impact of future land cover changes on HNO_3 and O_3 surface dry deposition.

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Abstract

Dry deposition is a key component of surface-atmosphere exchange of compounds, acting as a sink for several chemical species. Meteorological factors, chemical properties of the trace gas considered and land surface properties are strong drivers of dry deposition efficiency and variability. Under both climatic and anthropogenic pressure, the vegetation distribution over the Earth has been changing a lot over the past centuries, and could be significantly altered in the future. In this study, we perform a modeling investigation of the potential impact of land-cover changes between present-day (2006) and the future (2050) on dry deposition ~~rates~~velocities at the surface, with special interest for ozone (O_3) and nitric acid~~ vapor~~ (HNO_3), two compounds which are characterized by very different physico-chemical properties. The 3D chemistry transport model LMDz-INCA is used, considering changes in vegetation distribution based on the three future projections RCPs 2.6, 4.5 and 8.5~~.~~, and present-day (2007) meteorology. The 2050 RCP 8.5 vegetation distribution leads to a rise by up to 7% (+0.02 cm/s) in ~~VdO3~~the surface deposition velocity calculated for ozone (V_{d,O_3}) and a decrease of -0.06 cm/s in ~~VdHNO3~~the surface deposition velocity calculated for nitric acid (V_{d,HNO_3}) relative to the present day values in tropical Africa, and up to +18% and -15% respectively in Australia. When taking into account the RCP 4.5 scenario, which shows dramatic land cover change in Eurasia, ~~VdHNO3~~V_{d,HNO_3} increases by up to 20% (annual-mean value) and reduces ~~VdO3~~V_{d,O_3} by the same magnitude in

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this region. When analyzing the impact of surface dry deposition change on atmospheric chemical composition, our model calculates that the effect is lower than 1 ppb on annual mean surface ozone concentration, for both ~~for~~ the RCP8.5 and RCP2.6 scenarios. The impact on HNO$_3$ surface concentrations is more disparate between the two scenarios, regarding the spatial repartition of effects. In the case of the RCP 4.5 scenario, a significant increase of the surface O$_3$ concentration reaching locally by up to 5 ppb (+5%) is calculated on average during the June-August period. This scenario induces also an increase of HNO$_3$ deposited flux exceeding locally 10% for monthly values. Comparing the impact of land-cover change to the impact of climate change, considering a 0.93°C increase of global temperature, on dry deposition velocities, we estimate that the strongest increase over lands occurs in the North Hemisphere during winter especially in Eurasia, by +50% (+0.07 cm/s) for ~~VdO3~~V_{d,O_3} and +100% (+0.9 cm/s) for ~~VdHNO3~~V_{d,HNO_3}. However, different regions are affected by both changes, with climate change impact on deposition characterized by a latitudinal gradient, while the land-cover change impact is much more heterogeneous depending on vegetation distribution modification described in the future RCP scenarios. The impact of long-term land-cover changes on dry deposition is shown to be ~~non-negligible~~significant and to differ strongly from one scenario to another. It should ~~be~~ therefore be considered in biosphere-atmospheric chemistry interaction studies in order to have a fully consistent picture.

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1. Introduction

Amongst surface-atmosphere interactions, dry deposition plays a key role in the exchange of compounds and acts as a significant sink for several atmospheric ~~compounds~~species. Performing an intercomparison of 26 state-of-the-art atmospheric chemistry models, Stevenson et al. (2006) estimated the surface removal of ozone by dry deposition to be about 1000±200 Tg/yr on average, with values ranging from 720 to 1507 Tg/yr amongst models, compared to 5100, 4650 and 550 Tg/yr for chemical production, chemical destruction and stratospheric input fluxes respectively. This study also underlined that although global deposition fluxes are consistent between models, locally, there is a large variability in the ozone deposition velocities (Stevenson et al., 2006). Since all these models use deposition schemes based on Wesely's prescription~~,~~ (Wesely et al., 1989), the discrepancies suggest different hypotheses for the land-type consideration. Based on satellite measurements from OMI (Ozone Monitoring Instrument) combined with the Goddard Earth Observing System chemical transport model (GEOS-Chem), Nowlan et al. 2014 estimated dry deposition to land to be 98% of total deposition for NO_2 and 33% for SO_2. This deposition fluxes over land represent 3% of global NOx emissions and 14% of global ~~S~~sulfur emissions. Land surfaces can therefore play a significant role on deposition, with a ~~higlhy~~highly variable contribution~~ depending on the chemical compound considered /~~ from one chemical compound to another.

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The air-surface exchange of trace compounds has been shown to be strongly variable, especially between different types of surface vegetation and soil characteristics (Wesely et al., 2000). Regarding ozone, model data differences reported in the literature could be attributed to oversimplifications in the implementation of the dry deposition scheme (Val Martin et al. ~~2014~~2014) since many models rely on “resistance in series” schemes developed in the 1980s (Hardacre et al. 2005).

In order to quantify the non-photochemical sink for tropospheric burden at the regional and global ~~scale~~scales, the scientific community ~~has developed~~uses numerical dry deposition schemes calibrated with field-measurements of dry deposition velocities (Wesely et al., 1989, Zhang et al., ~~2002~~2002b), implemented usually in chemistry-transport models. Dry deposition efficiency is influenced by multiple meteorological factors (temperature, solar radiation, humidity and especially atmospheric turbulence), chemical properties of the trace gas considered (solubility, oxidative capacity), and land surface properties (surface type, surface roughness, foliar surface and ecosystem height in the case of vegetation surfaces). Some of these factors are poorly constrained and are thus accounted for in deposition schemes in ~~an~~a very simplistic way. The vegetation distribution for instance is usually prescribed using maps for the region of interest that are ~~usually~~generally kept the same for either past, present or future studies (e.g. Andersson and Engardt, 2010 or Lamarque et al. 2013).~~.~~ There is therefore a lack of knowledge regarding the impact of long-term changes in vegetation distribution on dry deposition chemical compounds at the surface. ~~Dry deposition parameters (resistances, fluxes, etc.) being especially tricky to measure, it limits the evaluation of dry deposition schemes, especially at the global scale for which a variety of surface and meteorological conditions should be documented. And yet, since the beginning of~~Since the beginning of the industrial era, human activities have modified the use of large surfaces, affecting significantly the vegetation distribution, especially in the northern temperate latitude regions. ~~Land~~Further land cover modifications are expected in the 21st century,

Mis en forme : Couleur de police : Automatique due to projected increases in energy and food demands~~. Vegetation~~, and vegetation, in tropical regions in particular, could undergo drastic alterations.

Only a few studies have been carried out recently on the dry deposition changes in the future. Some of them focus on the impact of climate change on the dry deposition (Andersson and Engardt, 2010) while others combine the effects of several future changes (climate, CO_2 levels, land cover) on atmospheric chemistry in general (Ganzeveld et al. 2010, Wu et al. 2012). However, considering anthropogenic land cover changes among other large modifications of the vegetation/atmospheric chemistry drivers does not allow to identify if the land cover change should be or not considered as a priority in the studies of future atmospheric chemistry. The objective of this study is to investigate and isolate the potential impact of land-cover changes between the present-day (2006) and the future (2050) on dry deposition ~~rates~~velocities at the surface, using a modeling approach with a 3D chemistry transport model as illustrated in Figure 1. Changes in vegetation distribution are based on the three future projections known as Representative Concentration Pathways scenarios (RCPs) (van Vuuren et al. 2011), developed for the climate model intercomparison project (CMIP5): RCPs 2.6, 4.5 and 8.5. For this work we focus on ozone (O_3) and nitric acid ~~vapor~~ (HNO_3), two compounds which are characterized by very different biophysical properties (e.g. solubility and ~~oxidant~~oxidative capacity). In section 2, we describe the chemistry-transport model LMDz-INCA, the dry deposition module and the modeling strategy adopted. In section 3, we describe the different future land cover changes as given in the ~~3~~three RCP scenarios 2.6, 4.5 and 8.5, and explain their impacts on surface dry deposition velocities of ozone and nitric acid~~ vapor~~. Finally, the magnitude of land cover effects related to climate change on dry deposition velocities by 2050 is discussed.

2. Modeling set up

In our study, the global chemistry-climate model LMDz-INCA (Hauglustaine et al., 2004) is used to compute dry deposition. LMDz (v4) is an atmospheric general circulation model that simulates the transport of trace species. The model is run with 19 hybrid levels from the surface to 3hPa at a horizontal resolution of 1.85° in latitude and 3.75° in longitude. It is coupled on-line to the chemistry and aerosols model INCA (v2) which computes concentrations of reactive tracers considering their emissions, chemical transformations, transport and deposition processes. The atmospheric oxidation reactions of CH_4, CO and non-methane hydrocarbons are documented in

Folberth et al. (2006). ~~Emissions~~In order to be able to isolate the effect of land-cover change only on the atmospheric chemical composition, through change in surface dry deposition, emissions are prescribed according to Lamarque et al. (2010) for anthropogenic fluxes and Lathière et al. (2006) for biogenic VOCs, as described in Szopa et al. ~~(~~(2013), and are kept constant between all runs.~~2013).~~

2.1 Dry deposition in LMDz-INCA

The chemical deposition scheme used in INCA is based on the parameterization of Wesely (1989) and Wesely and Hicks (2000), computing dry deposition velocity V_d as a succession of resistances as follows:

|V_d~~|=[~~(z)|=[R_a~~+~~(z)+R_b+R_c]$^{-1}$

where R_a is the aerodynamic resistance~~ (influenced by atmospheric stability and wind speed above the canopy),~~, R_b ~~is ~~the quasi-laminar resistance~~ (controlled by molecular diffusivity),~~, and R_c ~~is ~~the bulk surface resistance~~ (depending on both~~.

R_a determines the ability of the airflow to bring gases or particles close to the surface~~ and trace gas properties). ~~

~~The surface resistance R_c especially depends on specific surface (roughness length) [*Wesely, 1989*]) and meteorological (temperature and solar radiation) parameters. The surface occupancy type (presence of urban sites, water, vegetation, etc.) strongly modulates those parameters. In particular vegetation surfaces~~, and depends mainly on the atmospheric turbulence structure and on the height considered. In this paper, we will focus on dry deposition at the surface, ground level (z=0). R_b describes the resistance to the transfer very close to the surface and is driven by the surface (surface roughness) and the gas or particle (molecular diffusivity) characteristics. R_a and R_b are calculated based on Walcek et al. (1986). The surface resistance R_c represents the different pathways through which the gas or particles can deposit and is determined by the affinity of the surface for the chemical compound. Deposition can thus occurs directly on the ground and/or, in the case of vegetative surfaces, on the different vertical layers of the canopy on trunks, branches and mainly on leaves, through stomata or cuticles (Wesely, 1989). Vegetation surfaces in particular cover a large area of the Earth, with a high spatial and seasonal variability due to species diversity~~ and functioning, and are a key factor in dry deposition determination. ~~. Environmental conditions such as atmospheric CO_2 or pollutant (ozone) concentrations,

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radiation, temperature, or the occurrence of possible stress (drought for instance) can strongly affect the vegetation functioning, and the stomatal opening especially, and therefore impact dry deposition velocity. The impact of vegetation type, distribution and functioning, on dry deposition is still not well understood and generally very simply, if at all, considered in chemistry-transport models (Hardacre et al., 2015). For all chemical species considered in LMDz-INCA, R_c is based on their temperature dependent Henry's Law effective coefficient and reactivity factor for the oxidation of biological substances (Folberth et al., 2006). The coefficients for Henry's Law are taken from Sander (1999) and reactivity factors are taken from Wesely (1989) and Walmsley and Wesely (1996).

The dry deposition scheme implemented in LMDz-INCA considers eleven surface categories: (1) urban land, (2) agricultural land, (3) range land, (4) deciduous forest, (5) coniferous forest, (6) mixed forest including wetland, (7) water, both salt and fresh, (8) barren land, mostly desert, (9) non-forested wetland, (10) mixed agricultural and range land, and (11) rock open areas with low-growing shrubs. This scheme was originally developed by Wesely (1989) and updated by Wesely and Hicks ~~[~~(2000~~]~~) for Northern hemisphere regions of United States and southern Canada regions. Five seasonal categories are used as proxy of vegetation growth stage (midsummer with lush vegetation; autumn with unharvested cropland; late autumn after frost, no snow; winter, snow on ground, and subfreezing; transitional spring with partially green short annuals). For global scale study purposes, the scheme in LMDz-INCA has been modified in order to represent the different seasonal cycles throughout the world. The latitude dependency of the vegetation seasonality is described by dividing the globe into three belts: Northern hemisphere regions (latitude > 33°N); Tropical regions (33°S < latitude < 33°N) and Southern hemisphere regions (latitude < 33°S). Summer is considered in the tropics throughout the whole year, describing the evergreen vegetation. Two opposite seasonal cycles are taken into account in extra-tropical Northern and Southern hemisphere regions, with winter being activated when snow falls. The deposition of atmospheric compounds on plant leaves, through stomata especially, is determined following the Wesely (1989) approach. The stomatal resistance depends on vegetation type, seasonal category, radiation and temperature, but the potential impact of other environmental conditions such as drought, or atmospheric concentration of CO_2 or ozone, are not considered. The dry deposition velocity over each grid box is eventually determined by summing deposition velocities computed over every land cover types, weighted by their respective fractional surface coverage (ranging from 0 to 1).

The deposition ~~rates~~velocities computed by LMDz-INCA ~~are~~ based on a different land cover distribution was evaluated in Hauglustaine et al. (2004). This work illustratesvalues generally consistent with typical deposition velocities ~~exposed~~highlighted for North America and Europe as presented in Wesely and Hicks~~,~~ (2000) and ~~more generally with global models with~~ monthly values reaching up to 0.6 cm~~.~~/s~~$^{-1}$~~ for ozone and up to 3 cm~~.~~/s~~$^{-1}$~~ for HNO_3 over land. In the supplementary material the ozone dry deposited fluxes simulated by LMDz-INCA in the present-day simulation and used in this study are compared to other global model and long term measurements which are discussed in Hardacre et al. (2015).

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2.2 Land use and land cover changes between 2007 and 2050

The present-day distribution of vegetation categories considered in LMDz-INCA is illustrated in Figure ~~1~~2 as dominant type, covering the largest fraction of each gridbox. Crops are dominant mainly in restricted temperate regions of North America, Central Europe, and also in India, while range lands are largely spread. Deciduous forests dominate in tropical regions of South America, Africa and Indonesia, together with Central and Southern Europe, while coniferous forests have a high occupancy in boreal regions of North America and Eurasia. ~~The~~ Figure ~~1~~2 also shows the 10 regions of special interest selected for this study, ~~that~~which will be considered in more ~~details~~detail when analyzing our results.

Future maps are based on scenarios of land-cover changes derived from four different Representative Concentration Pathways (RCPs; Moss et al. 2010; Van vuuren et al. 2011) and four Integrated Assessment Models (one per RCP) (RCP 8.5, RCP 4.5 and RCP 2.6). Those maps were further harmonized to ensure smooth transitions with past/historical changes (Hurtt et al. 2011). Those datasets only provide information on human activities (crop land and grazed pastureland) in each grid-cell (at a 0.5° resolution) but do not provide any recommendation regarding the distribution of natural vegetation. We have therefore combined them with our original present-day land-cover map (Loveland et al. 2000), which already includes both natural and anthropogenic vegetation types, following a methodology described in Dufresne et al. (2013). Figure ~~2~~3 illustrates changes in vegetation fraction for agriculture and grasslands on one hand, and for forests on the other hand, between present-day (distribution for 2007) and the future RCP scenarios. For most affected regions, the changes in land surfaces are presented in Figure ~~3~~4. The RCP 4.5 scenario shows the largest surface change with a total of 20.8 x 10^6 km^2, representing 10.4% of the 70°S-70°N Earth continental surface. According to the RCP 2.6 and RCP 8.5 scenarios only 15 to 16.8 x 10^6 km^2 of land cover surfaces are converted.

The RCP 2.6 scenario is characterized by a moderate increase of energy consumption throughout the 21st century together with a decrease in oil consumption. The energy supply is thus partly ensured by bioenergy production increase (van Vuuren et al, 2011). Such hypotheses lead to a strong expansion of agricultural lands (+ 2.61 x 10^6 km^2 globally) at the expense of forests (- 1.40 x 10^6 km^2) and grasslands (- 1.15 x 10^6 km^2) targeting mainly Eurasia, US and tropical southern America.

The RCP 8.5 scenario, characterized by the strongest increase in population and energy consumption~~ (~~, amongst RCPs), assumes a large increase in global population until 2050. The resulting demand for food leads to a strong expansion of land used for crops and pastures at the expense of forests. The tropical belt (from 30°N to 30°S) undergoes the largest changes: tropical forests in southern America and southern Africa are partially harvested (1.0 x 10^6 km^2 totally, i.e. 13% of their 2007 extent) and replaced by grassland and crops, while in Eastern Australia, forests lose 7% (- 0.28 x 10^6 km^2) of their 2007 area and are replaced by grasslands which gains 0.12 x 10^6 km^2 on desert.

The “mitigation” RCP 4.5 scenario is a rather contrasting scenario as it proposes a strong increase in ~~all forests’ occupancy~~the cover of all forest categories, a small expansion of grasslands but an important recession of agricultural surfaces mainly in developed countries. Indeed Eurasia~~ and~~, US and Canada undergo a strong conversion from agriculture and grassland to forests with a magnitude change of ~0.8 x 10^6 km^2 in Eurasia and ~0.4 x 10^6 km^2 in northern US and Canada. Besides, tropical southern America ~~losses~~loses 0.55 x 10^6 km^2 of cumulated croplands and grasslands but forests expand by the same surface between present day and 2050.

Finally, it is important to underline that the 3 RCP scenarios offer a wide variety of land cover change projections. They all are quite different compared to previous scenarios, such as the SRES-A2 investigated by Ganzeveld et al. (2010), characterized by a strong North/South contrast, with the tropical and southern hemisphere countries mainly encountering deforestation whereas northern areas (>35°N) were mainly projected to see afforestation.

2.3 Simulation strategy

In order to quantify the effects of these land cover changes on surface dry deposition, we carried out two sets of simulations (Table 1). The first set intends to isolate the effect of future possible land cover changes on dry deposition ~~prior to~~without any climate change. It includes one control run (present day), using 2006 vegetation distribution (Figure ~~1~~2) and three future runs using the 2050 vegetation maps according to the RCPs 8.5, 4.5 and 2.6 scenarios. The same present-day meteorology, biogenic and anthropogenic emissions are used in these four simulations. These simulations are run for 1 year with wind and temperature fields being relaxed towards the ECMWF ERA-interim reanalysis (Dee et al., 2011) with a time constant of 6 hours.

Then a second set of two simulations is performed in order to investigate the effect of future climate change on deposition and compare it with the impact of future land cover change: one run for the 2000-2010 period and a second run for the 2045-2055 period. Those simulations are performed without nudging and the LMDz general circulation model requires sea surface temperature (SST), solar constant and Long-Lived Green House Gases (LL-GHG) global mean concentrations as forcings. For historical simulations, we use the HADiSST for sea surface temperature (Rayner et al., 2003) and the evolution of LL-GHG concentrations compiled in the AR4-IPCC report. For future projections, we use the SST from IPSL-CM4 simulation for the SRES-A2 scenario, which induce similar climate trajectories in terms of radiative forcing than RCP8.5. We use the LL-GHG concentrations distributed by the RCP database for RCP8.5 projection for the 2045-2055 period. Eleven years are run and averaged to allow smoothing of interannual climate variability. The mean surface temperature ~~changes~~change is 0.93°C between future simulation and present day simulation. Both experiments use the same present-day vegetation distribution, anthropogenic and biogenic emissions.

3. Results

3.1. Present day ozone and nitric acid ~~deposition~~

~~First~~**deposition**First of all, we present the deposition over continental regions for present-day conditions (Figure ~~4~~5) by illustrating the annual means of deposition ~~rates~~velocities at the surface, surface concentrations and deposited fluxes for O_3 and HNO_3.

The highest ozone deposition velocities (>0.35 cm~~.~~/s~~-1~~) are simulated over India, south-eastern Asia, western coast and center of ~~south~~South America, Mexico, Europe and sub ~~saharian~~Saharan

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Africa and Australia. Hence, those areas are mainly covered by crops and grasses, where the highest ~~VdO3~~V_{d,O_3} occurs, while Europe and Southeast Asia are mainly covered by deciduous forests, with therefore lower ~~VdO3. O3~~annual V_{d,O_3}. O_3 surface dry deposition is indeed maximal over small canopies vegetation and minimal over bare soil with deposition affinity ranging from Agriculture > Grasslands > Deciduous > Coniferous > Bare soil (see sensitivity tests in supplementary material).

Temperate regions see ozone deposition velocities significantly reduced in winter (see supplementary material for ~~monthly~~seasonal means) whereas tropical regions, covered mainly by small canopies, are characterized by surface deposition ~~rate~~velocity exceeding 0.35 cm~~.~~/s~~-1~~ throughout the whole year due to the lack of seasonality in the vegetation phenology in the global model. In temperate regions of the Northern hemisphere, the highest ~~values of~~ deposition ~~velocity~~velocities for ~~both~~ ozone ~~reaching monthly mean~~reach values of 0.4 cm/s to 0.6 cm/s for ~~VdO3~~V_{d,O_3} over Europe.

For HNO_3, the annual mean deposition ~~rates~~velocities are maximum over Brazil, Western Europe, India, Indochinese Peninsula and South of western Africa (> 1.~~6cm. -1~~6 cm/s in annual mean). ~~VdHNO3~~V_{d,HNO_3} reaches maximum values over deciduous and coniferous forests, due to deposition affinity ranking from: Deciduous, Coniferous > Agriculture > Grasslands > Bare soil. This is due to the strong dependency of ~~VdHNO3~~V_{d,HNO_3} to surface roughness (Walcek et al., 1986). For temperate region and South Asia, the HNO_3 deposition is strongly affected by the vegetation cycle with maximum in July between 2.5 cm/s and 3.5 cm/s. This is remarkable over temperate and boreal forests. In the tropics, Amazonian forest encounters high HNO_3 deposition ~~rate~~velocity in ~~January~~winter whereas deposition ~~rate~~velocity over African equatorial forest is limited throughout the whole year. ~~(see supplementary material for monthly means of deposition).~~(see supplementary material for seasonal means of deposition). Large areas receive high HNO_3 deposition fluxes exceeding 0.5g(N)/m^2/yr in annual mean: North eastern USA, western Europe and East Asia. These areas correspond to the ones identified by Dentener et al. 2006 and in which natural vegetation encounters nitrogen deposition higher than the “critical load” threshold of 1g(N)/m^2/yr.

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The repartition of deposited fluxes is strongly affected by the large variability of atmospheric concentrations of ozone and nitric acid in the surface layer. For both O_3 and HNO_3, the deposited fluxes are maximum over South and East Asia and eastern North America and central and

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western Europe. For ozone, the maximum in ~~January~~winter is over central Africa whereas in ~~July~~summer the ozone deposition is maximum over central Europe and eastern US. For HNO_3, the deposited flux repartition ~~mixes more~~is equally driven by the deposition ~~rate~~velocity and by the HNO_3 surface ~~concentrations heterogeneities.~~concentration distribution. In ~~January~~winter, HNO_3 is maximally deposited over eastern US, central Africa, central Europe India and East Asia. In ~~July~~summer, regions are the same in the Northern hemisphere but the extension of deposited HNO_3 areas is higher and the deposition in Africa is weak, due to weak HNO_3 concentration.

3.2. Impact of 2050-2007 land cover changes on surface dry deposition velocities.

We then analyze the changes in surface dry deposition velocities between present day and 2050 induced only by land cover ~~changes~~change. Four regions undergo interesting land cover changes in ~~term~~terms of intensity or contrast between scenarios: Eurasia, North America, tropical Africa and Australia. The left columns of Figures ~~5~~6 and ~~6~~7 show the relative difference in surface dry deposition velocities distribution for O_3 and HNO_3, resulting from the changes in vegetation distribution between 2007 and 2050 for the 3 RCP scenarios. We shall first describe the two scenarios projecting weak land cover changes for 2050s: RCP8.5 and RCP2.6. In the RCP 8.5 scenario, one main land cover change is the expansion of agricultural land at the expenses of forests. According to this scenario, over tropical Africa, the maximal land cover change occurs locally with fraction of deciduous forests decreasing by up to 0.2 while cropland fraction increases by up to 0.2 in the same region. This induces a rise by up to 7% (+0.02 cm/s) in ~~VdO$_3$~~V_{d,O_3} and a decrease of -0.06 cm/s in ~~VdHNO$_3$~~V_{d,HNO_3} relative to the present day values in this area. These order of magnitude and sign of changes are consistent with sensitivity tests in which we replaced totally forests by croplands inducing an increase of 0.1 cm/s in ~~VdO$_3$~~V_{d,O_3} and a decrease of 0.5 cm/s in ~~VdHNO$_3$~~V_{d,HNO_3} (during summer and winter). The strongest LCC occurs in Australia (-0.12 in forest fraction and +0.2 in grassland fraction in eastern Australian regions), which induces a local maximum increase of 18% (+0.05 cm/s) in ~~VdO$_3$~~V_{d,O_3} and a maximum decrease of 15% in ~~VdHNO$_3$~~V_{d,HNO_3} (-0.1 cm/s). We find the same order of magnitude in changes induced by land cover change in Western Australia but with a different sign for

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~~VdHNO$_3$~~V_{d,HNO_3} changes (+0.1cm/s ; +9%), due to a different type of shift in surface covering (+0.12 in grassland fraction, -0.10 for desert).

As land cover changes are weak in the RCP 2.6 scenario, a more dispersed and weaker effect on surface dry deposition velocities is simulated (maximum absolute difference of 10%).

According to the RCP 4.5 scenario, the most dramatic land cover change occurs in Eurasia where local maximum changes ~~of~~by up to 0.5 in fraction of vegetation are projected, involving in most cases an increase in forest ~~surface~~surfaces at the expense of agricultural ~~area~~areas. This increases ~~VdHNO3~~V_{d,HNO_3} by up to 20% (annual-mean value) and reduces ~~VdO3~~V_{d,O_3} by the same magnitude in this region. The LCC impacts are stronger by a factor 4 to 6 in summer both on O_3 and HNO_3 deposition ~~rates~~velocities. This difference in deposition velocities between winter and summer were highlighted in sensitivity tests which see a strong decrease in ~~VdO3~~V_{d,O_3} during the June-August period (up to 0.15 cm/s in absolute) and a strong increase in ~~VdHNO3~~V_{d,HNO_3} (up to 1.5 cm/s) underlining a total conversion of croplands to forests. This is due to a higher surface roughness which enhances the deposition velocity of HNO_3 (via the reduction of the aerodynamic resistance). However, the higher input surface resistance (prescribed in the model and variable relating to season indexes) reduces ~~VdO3~~V_{d,O_3} even combined to a warmer climate which decreases the stomatal resistance (Rs).

3.3. Impact on atmospheric composition

The objective of this part is to isolate the effects of dry deposition changes due to land cover changes on the tropospheric concentration of O_3 and HNO_3. Therefore, solely the impact of land cover changes on deposition at the surface is considered between the present-day and 2050 simulations. This impact on surface concentrations of O_3 and HNO_3 is shown in the right columns of Figures ~~5~~6 and ~~6~~7.

For both the RCP8.5 and RCP2.6 scenarios, the ~~LLC~~LCC effects through deposition are lower than ~~1ppb~~1 ppb on annual mean surface ozone concentrations. In term of relative difference, only the reduction of ozone over Australia when considering RCP8.5 hypotheses is exceeding 1%, reaching up to 5% at some points. The impact on HNO_3 surface concentrations is more disparate between the two scenarios when considering the spatial repartition of effects. The RCP8.5 ~~leads~~scenario leads to local increase of HNO_3 due to the reduction ~~of~~in the deposition ~~rate~~velocity. This HNO_3 increase is notable over Mexico, Brazil, western and South Africa (comprised in the

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1-6% interval). Land cover change in Australia leads to an increase exceeding 7% ~~on~~in the ~~eastern part~~east and a decrease reaching 5% ~~on~~in the ~~western part~~west.

The RCP4.5 scenario induces the strongest impacts on deposition ~~rate~~velocity with a reduction of ~~VdO3~~V_{d,O_3} (-0.08 cm/s) occurring in Eurasia due a strong reduction in croplands occupancy (-0.6 in fraction of coverage) and a strong increase in forest distribution (+0.6 in fraction of coverage) between 2007 and 2050. It induces a significant increase of the surface O_3 concentration reaching locally by up to 5 ppb (+5%) on average during the June-August period. This scenario induces also an increase of the HNO_3 ~~deposited~~deposition flux exceeding locally 10% for monthly values. In Eurasia and eastern North America. It thus leads to a reduction in the HNO_3 concentration ~~of~~by 0.2 ~~ppb~~ppbv in Eurasia (-13%) and in North America (-8~~%) via~~%), mainly due to changes in nitric acid ~~vapor~~ velocities of +0.5 cm/s and +0.2 cm/s respectively.

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3.4 Are the land cover induced changes significant compared with the climate change impact?

The impact of land-use changes on deposition can be compared to the one of climate in order to discuss their respective strength on deposition ~~rates. In~~velocities. To this purpose, we consider a 0.93°C increase of global temperature, corresponding to the temperature increase ~~planned~~projected in the RCP ~~projections~~scenarios between the beginning and the middle of the 21st century. The figure ~~7~~8 shows the impact of this climate change on the deposition ~~rate~~velocity for O_3 and HNO_3. We see that the strongest increase in surface dry deposition velocities over lands occurs in the North Hemisphere during winter especially in Eurasia (+50% (+0.07 cm/s) for ~~VdO3~~V_{d,O_3} and +100% (+0.9 cm/s) for ~~VdHNO3~~V_{d,HNO_3}). The climate effect on the deposition ~~rate~~velocity by affecting stomatal resistance, sensitive to surface temperature and solar ~~radiance~~irradiance, can locally reach values far more important than the LCC. The Table 2 presents the effects of land cover change considering RCP4.5 projection and climate change on deposition ~~rate~~velocity averaged over 10 regions for O_3 and HNO_3. In several regions, the effect of land cover change is of the same order of magnitude than the one of climate. The modification in land cover affectation can thus amplify the climate change effect or, when the sign is the opposite, counterbalances it.

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4. ~~Conclusion and~~ Discussion and conclusions

Using the 2.6, 4.5 and 8.6 RCP scenarios for land-use change between 2000s and 2050s, simulations were carried out with the global chemistry-transport model LMDz-INCA in order to assess ~~to~~the impact of changes in vegetation distribution on the dry deposition of ozone and nitric acid at the surface and on atmospheric composition.

Regarding vegetation distribution, the largest change at the global scale is given in the RCP 4.5 scenario (20.8 x 10^6 km²), with surface converted being 28% and 19% lower in the RCP 2.6 and RCP 8.5 scenarios respectively. Projections show major changes in the Northern Hemisphere in the case of RCP 4.5 scenario, while Australia and Africa are mostly affected in the RCP 8.5 scenario.

Vegetation type and surface being key drivers of surface dry deposition, any change in vegetation distribution can potentially affect dry deposition velocity and therefore atmospheric chemical composition. Considering the 2050 RCP 8.5 vegetation distribution leads to a rise by up to 7% (+0.02 cm/s) in ~~VdO$_3$~~V_{d,O_3} and a decrease of ~~-~~0.06 cm/s in ~~VdHNO$_3$~~V_{d,HNO_3} relative to the present day values in tropical Africa, and up to +18% and -15% respectively in Australia. As land cover changes are weak in the RCP 2.6 scenario, a more dispersed and weaker effect on surface dry deposition velocities is simulated (maximum absolute difference of 10%) when considering the RCP 2.6 scenario, characterized by a moderate change in vegetation distribution compared to present-day. When taking into account the RCP 4.5 scenario, which shows dramatic land cover change in Eurasia~~ VdHNO3,~~ V_{d,HNO_3} increases by up to 20% (annual-mean value) and reduces ~~VdO3~~V_{d,O_3} by the same magnitude in this region. When analyzing the impact of dry deposition change on atmospheric chemical composition, our model calculates that the effect is lower than 1 ppb at the grid box scale on annual mean surface ozone concentration, for both ~~for~~of the RCP8.5 and RCP2.6 scenarios. The impact on HNO_3 surface concentrations is more disparate between the two scenarios, regarding the spatial repartition of effects. In the case of the RCP 4.5 scenario, a significant increase of the surface O_3 concentration reaching locally up to 5 ppb (+5%) is calculated on average during the June-August period. This scenario induces also an increase of HNO_3 deposited flux exceeding locally 10% for monthly values. Investigating the impact of climate change, considering a 0.93°C increase of global temperature, on surface dry deposition velocities, we calculate~~ see~~ that the strongest increase over lands occurs in the North Hemisphere during winter especially in Eurasia (+50% (+0.07 cm/s) for ~~VdO3~~V_{d,O_3} and +100% (+0.9 cm/s) for ~~VdHNO3~~V_{d,HNO_3}). The climate change impact on deposition is characterized by a latitudinal

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gradient, while the effect of land-cover change is much more heterogeneous. Both climate and vegetation distribution changes are of similar amplitude but sign can differ.

The objective in this study is to isolate the impact of land-cover change on atmospheric chemical composition through modification of surface dry deposition only rather than to consider comprehensively all the atmospheric chemistry/vegetation interactions affected by land cover change. Indeed, as far as long term evolution of atmospheric chemistry is investigated (e.g. Stevenson et al. 2006, Lamarque et al. 2010), the evolution of biogenic emissions due to global changes is discussed, if not shared between models, but the land cover maps used for dry deposition remain unchanged. Here we want to assess the importance of this choice. Land cover changes would go together with changes in surface emissions, either from anthropogenic, agricultural, or biogenic sources, with changes in climate, and possible strong consequences on the atmospheric chemical mechanism and surface-atmosphere interactions. In an attempt to quantify all the effects of land cover change, those processes would therefore need to be considered altogether to get a better picture of the overall resulting effect. However they all have large uncertainties and added to error compensation effects, the dry deposition change can be can masked by other process changed (see for example Wu et al., 2012). Moreover, the sensitivity of biogenic emissions to climate and CO_2 changes as well as the level of coupling between vegetation and chemistry are so different from one model to another that the full land cover change response is for the moment highly model-dependent.

Fowler et al. (2009) underline an uncertainty of about 50% in the ability of models to estimate dry deposition fluxes for main chemical species, the lack of measurements making a proper and extensive model evaluation especially difficult. Hardacre et al. (2015), who compared the dry deposition of ozone of 15 global atmospheric chemistry-transport models with measurements in Europe and North America underline discrepancies of up to a factor of two, notably in the summer maximum, but do not find a systematic model bias. Dry deposition in global models is still largely based on the in-series resistance approach proposed by Wesely (1989) and generally do not integrate more recent findings demonstrated by field or laboratory studies (Hardacre et al., 2015).

Vegetation is usually crudely described in chemistry-transport models, with leaf surface or cuticle and stomatal resistances for instance being prescribed or very simply parameterized, and lack of the representation of seasonal variation or stress (water, temperature) impacts. This could lead to significant uncertainty in model representation and projections of atmospheric chemical composition and surface-atmosphere interactions. The work by Wesely and Hicks (2000) underlines that selecting proper input parameters ~~of~~for dry deposition ~~scheme~~schemes, such as stomatal, cuticle, and soil resistances, is crucial for a satisfactory determination of dry deposition efficiency, for both simple and multi-layers models. Zhang et al. (2003) propose a revised parameterization of dry deposition including the leaf area index in the calculation of aerodynamic and cuticular resistances, which could give the possibility of a better representation of the impact of vegetation seasonality in dry deposition estimates. The roles of surface wetness, soil moisture, the partition between stomatal and non-stomatal uptake for instance, shown of high importance for dry deposition processes, are usually not implemented or poorly described in global models (Fowler et al., 2009; Hardacre et al., 2015). This is also the case of the LMDz-INCA model in which dry deposition is described through a highly parameterized approach. Investigating ozone non-stomatal uptake using measurements over five different vegetation types, Zhang et al. (2002a) show that the O_3 uptake by cuticles is affected by friction velocity, relative humidity, canopy wetness and LAI especially, and tends to increase with wetness and high humidity. A new parameterization for non-stomatal uptake is proposed and is expected to improve this deposition path in existing models, where a constant value is often considered, and could therefore be tested more largely in global models. Investigating the impact of coupling dry deposition to vegetation phenology in the Community Earth System Model (CESM) on ozone surface simulation, Val Martin et al. (2014) ~~shows~~show the importance of representing the dependence of dry deposition to vegetation parameters including drivers of stomatal resistance variation (change in CO_2, drought stress), especially when focusing on the impact of past or future changes of vegetation. ~~Next~~Hardacre et al. (2015) recommend to provide more detailed diagnostics of O_3 dry deposition in next intermodel exercices to attribute the intermodal differences to methodology and/or representation of processes. The next generation of chemistry-transport models should therefore rely on online coupling with vegetation, with dry deposition schemes having a consistent and dynamic description of vegetation distribution and growth and related short-term (seasonal, annual variation) or long-term (past and future changes) evolutions. However, model intercomparisons focusing on each process considered in isolation with a proper shared methodology/set-up is crucial if one wants to progress in the understanding of the complex vegetation/atmospheric chemistry interactions. In particular the evolution of land cover maps should be considered as far as dry deposition is concerned in addition to emission changes in the next intermodel exercices aiming to project future atmospheric chemistry.

Acknowledgements

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Table 1 : Simulations performed in our study with the LMDz-INCA chemistry-climate model : set-up description.

Run objectives	**Land-cover map**	**Climate**	**Duration**
CONTROL	Present-day 2000s	Winds and surface temperature nudged on ECMWF fields for 2007	1 year
IMPACT OF FUTURE LAND-USE CHANGES	2050 RCP 8.5	Winds and surface temperature nudged on ECMWF fields for 2007	1 year
	2050 RCP 4.5		
	2050 RCP 2.6		
IMPACT OF FUTURE CLIMATE	Present-day 2000s	2000-2010 fields (GCM mode)	10 years
		2045-2055 fields (GCM mode)	

Table 2 : Mean Effect on Annual Mean Surface Deposition ~~Rate~~Velocity (%) of climate and land cover changes of O_3 and HNO_3 averaged over homogeneous regions (values > +-1.5% are highlighted)

	Ozone			Nitric Acid		
	Climate Change	RCP4.5 Land cover Change	Sum of Climate and Land cover Changes	Climate Change	RCP4.5 Land cover Change	Sum of Climate and Land cover Changes
GLOBAL	0.5	-0.7	-0.2	**2.2**	1.2	**3.4**
Eurasia	**2.1**	**-2.1**	0.0	**4.3**	**3.8**	**8.1**
USA	**1.5**	-1.3	0.2	**3.6**	**2.0**	**5.6**
Central America	-1.1	-1.4	**-2.6**	1.1	**1.7**	**2.8**
Tropical Southern America	**-2.3**	-1.2	**-3.5**	1.1	**2.6**	**3.7**
Tropical Africa	**-1.5**	-0.8	**-2.3**	0.4	0.9	1.3
South Africa	-1.4	-0.6	**-2.0**	-0.1	0.8	0.8
West Australia	-0.4	-0.1	-0.5	-0.4	0.0	-0.4
East Australia	-0.5	-0.6	-1.1	0.2	0.5	0.7
South America	0.4	-0.7	-0.4	0.3	**2.0**	**2.3**
Tropics	-1.1	-0.6	**-1.7**	0.6	1.0	**1.7**

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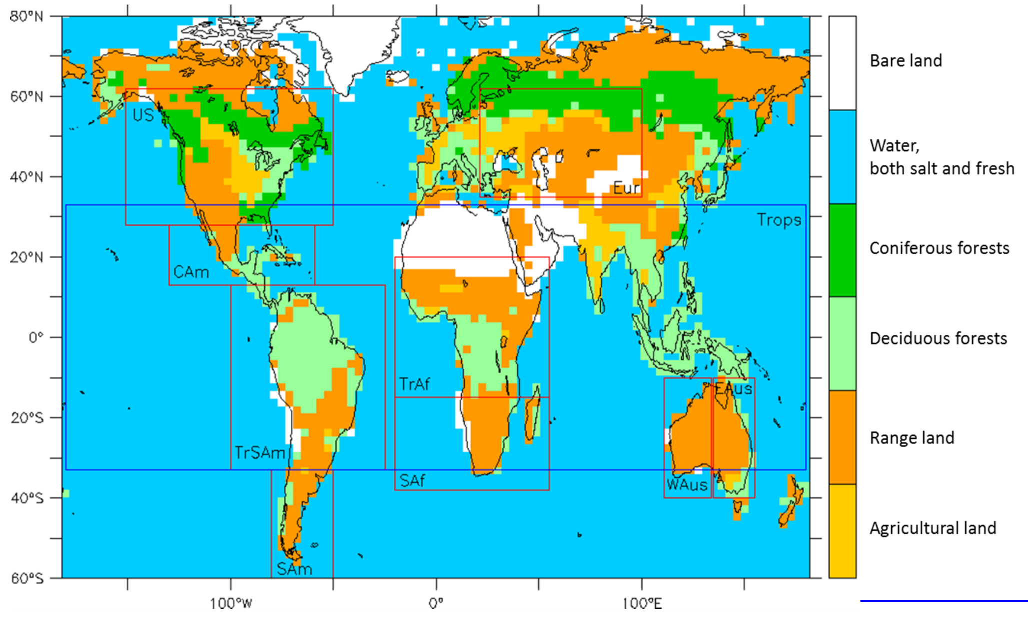

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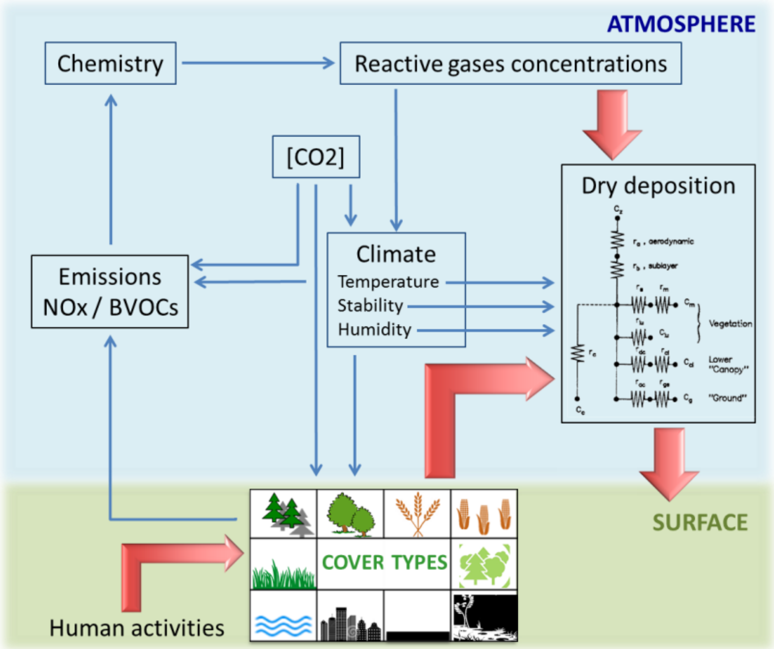

~~Figure 1~~

Figure 1: Interactions between vegetation and atmospheric chemistry potentially affected by land use changes. In this work, only the red arrows are investigated.

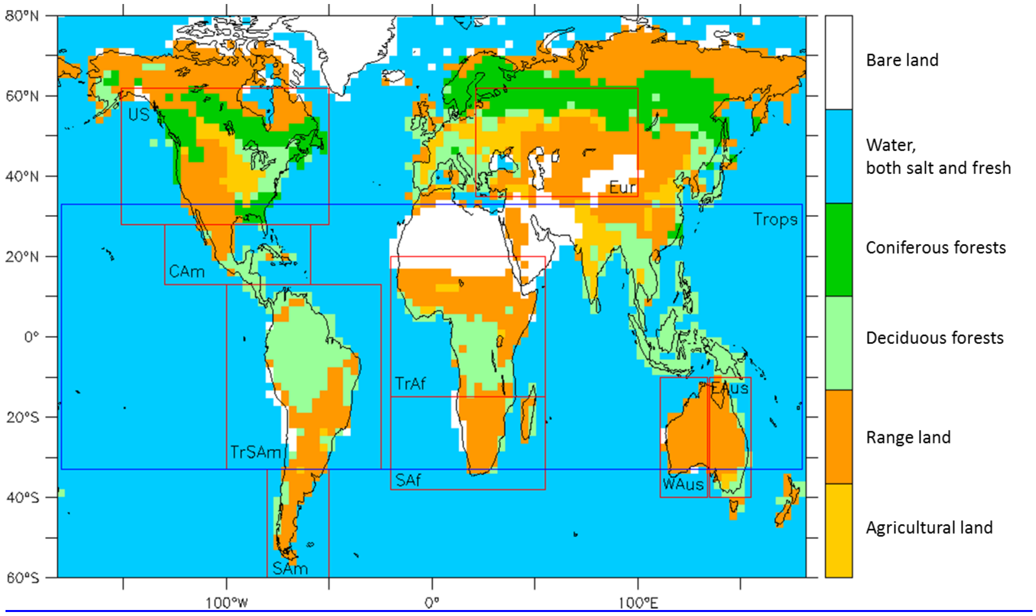

Figure 2: Surface categories considered in LMDz-INCA for dry deposition, represented as dominant coverage : agricultural land, range land, deciduous forest, coniferous forest, water, barren land, mostly desert. Regions discussed in this study are also illustrated: Eurasia, USA, Central America, Tropical Southern America, Southern America, Tropical Africa, Southern Africa, Western Australia, Eastern Australia and Tropical regions.

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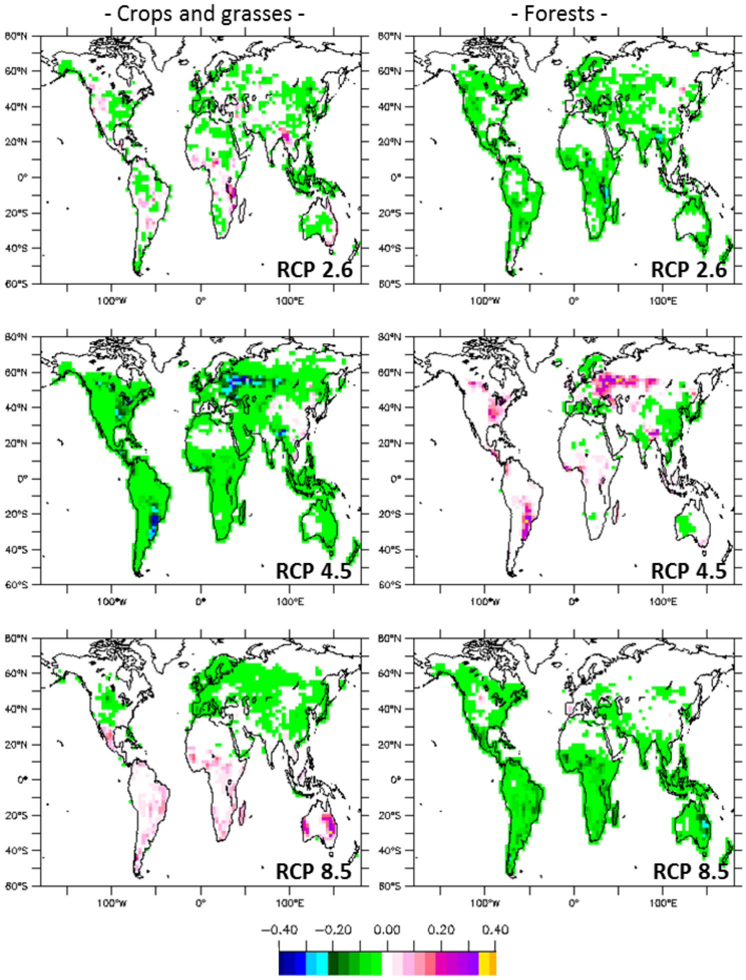
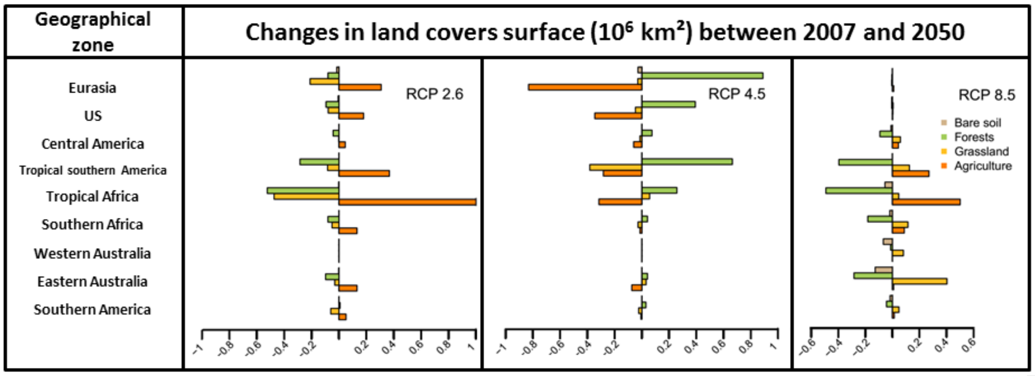


Figure ~~2~~3: Vegetation fraction difference between 2050 and present-day for crops and grasses (left column), and forests (right column) according to the future RCP scenarios 2.6 (upper line), 4.5 (middle line) and 8.5 (lower line).



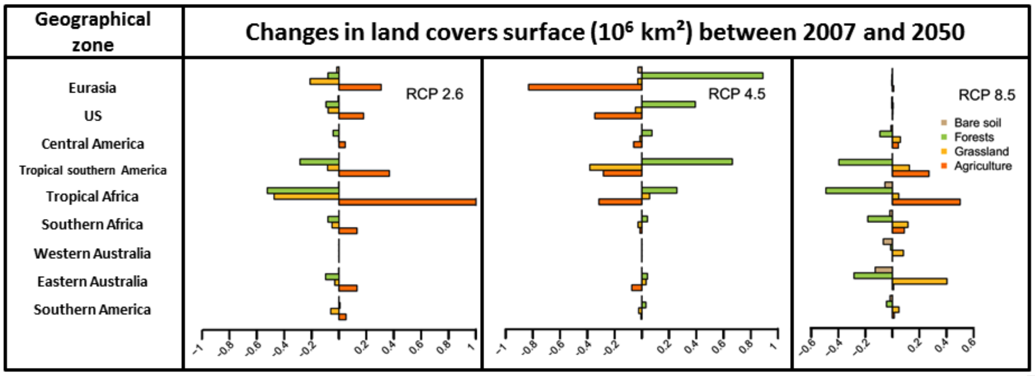
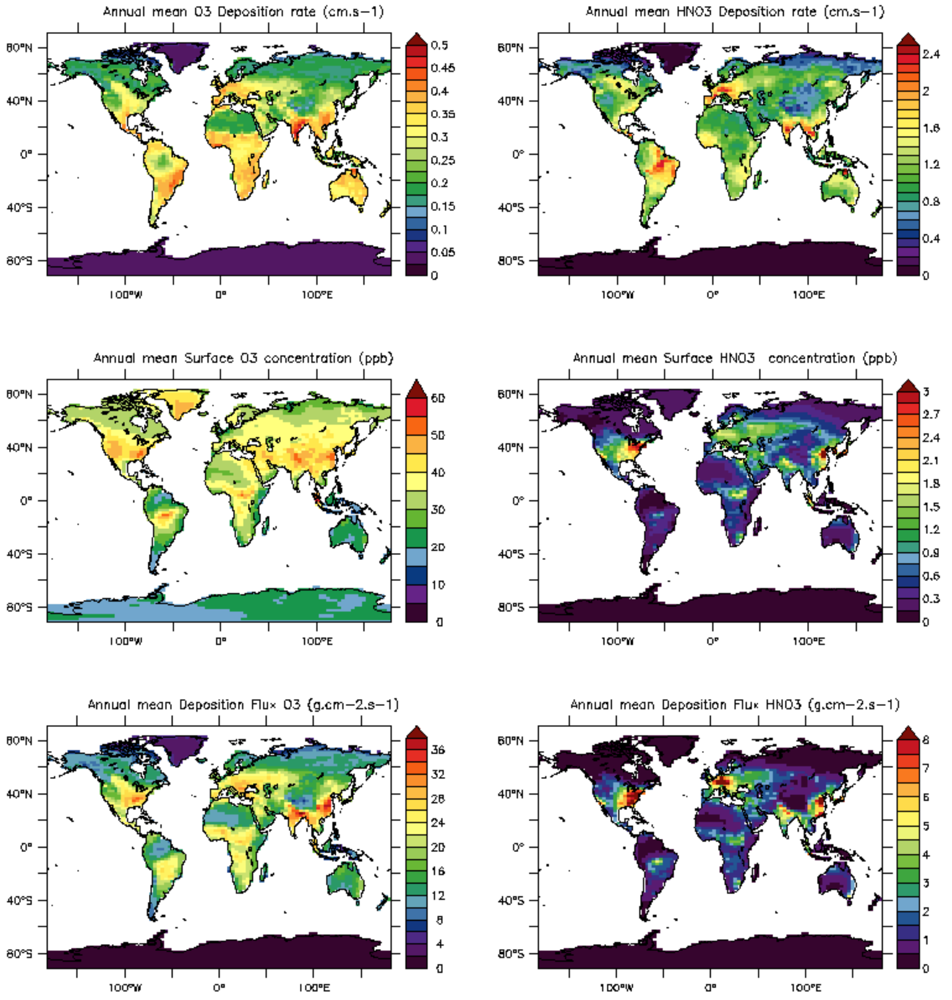


Figure ~~3~~4: Changes between 2007 and 2050 in land-type surfaces (10^6 km²) for the nine regions as illustrated in figure 1, in the case of forests (green), crops (orange), grasses (yellow) and bare soil (brown).


Annual mean O3 Deposition rate (cm.s−1)
0.5
0.45
0.4
0.35
0.3
0.25
0.2
0.15
0.1
0.05
0
Annual mean HNO3 Deposition rate (cm.s−1)
2.4
2
1.6
1.2
0.8
0.4
0
Annual mean Surface O3 concentration (ppb)
60
50
40
30
20
10
0
Annual mean Surface HNO3 concentration (ppb)
3
2.7
2.4
2.1
1.8
1.5
1.2
0.9
0.6
0.3
0
Annual mean Deposition Flux O3 (g.cm−2.s−1)
36
32
28
24
20
16
12
8
4
0
Annual mean Deposition Flux HNO3 (g.cm−2.s−1)
8
7
6
5
4
3
2
1
0
80°N
40°N
0°
40°S
80°S
100°W
0°
100°E

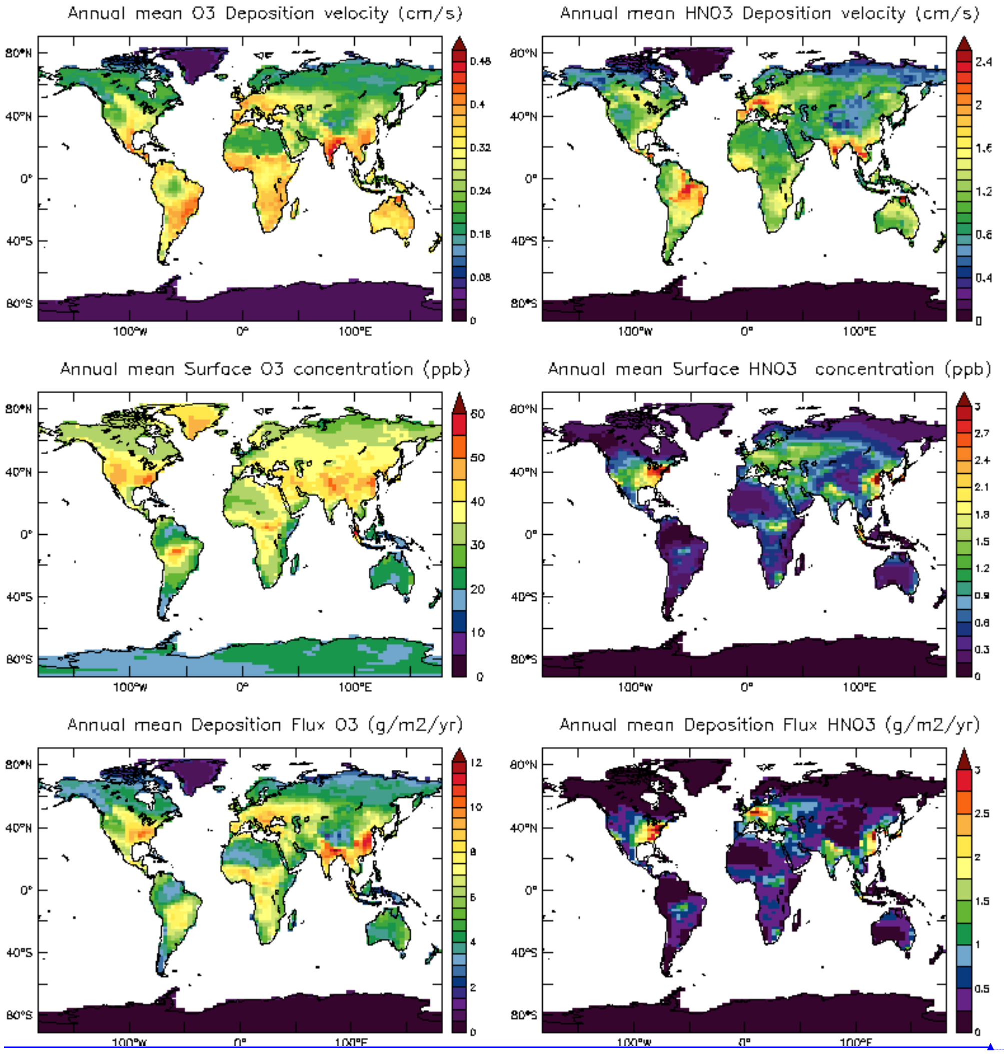


Figure ~~4~~5: Annual average of surface dry deposition velocities (upper panel), surface concentrations (middle panel and deposition fluxes (lower panel) over continental surfaces (cm/s) for O_3 (left) and HNO_3 (right) for present-day as simulated by LMDz-INCA.

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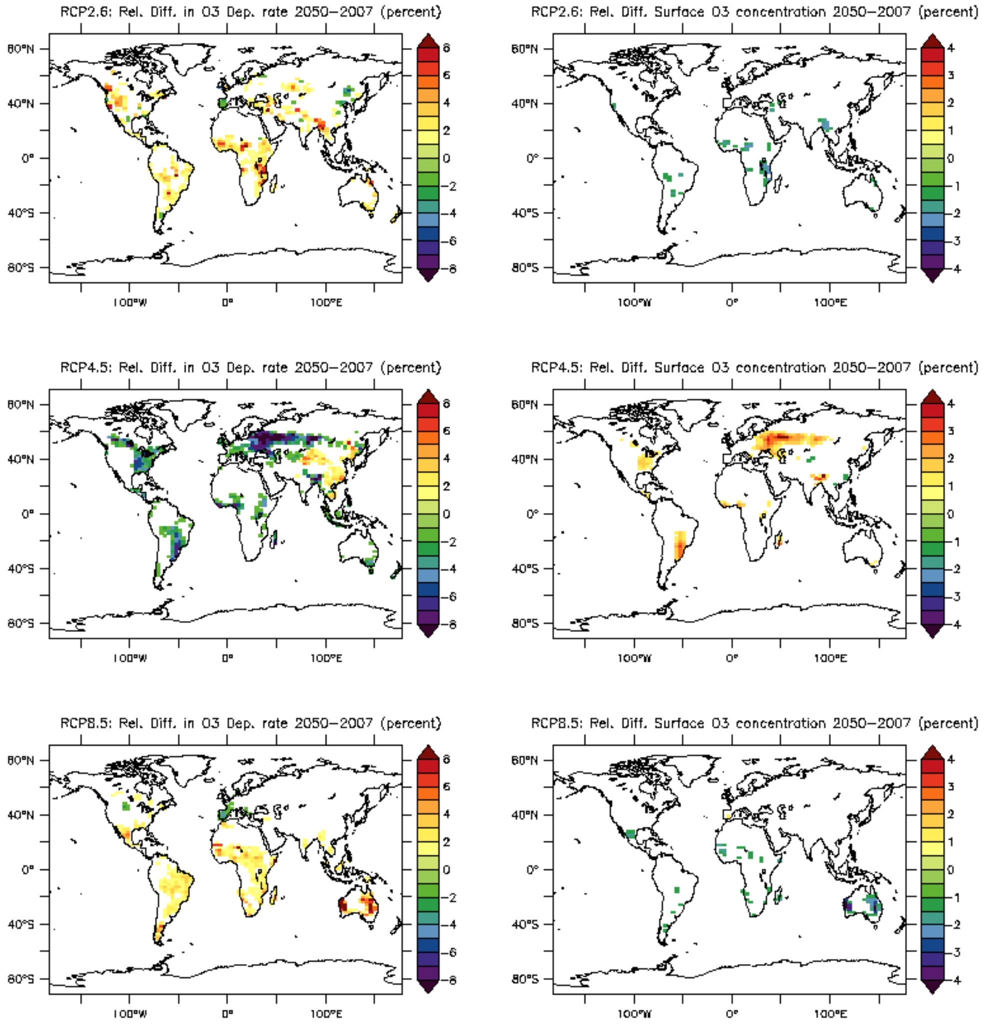


RCP2.6: Rel. Diff. in O3 Dep. rate 2050–2007 (percent)

RCP2.6: Rel. Diff. Surface O3 concentration 2050–2007 (percent)

RCP4.5: Rel. Diff. in O3 Dep. rate 2050–2007 (percent)

RCP4.5: Rel. Diff. Surface O3 concentration 2050–2007 (percent)

RCP8.5: Rel. Diff. in O3 Dep. rate 2050–2007 (percent)

RCP8.5: Rel. Diff. Surface O3 concentration 2050–2007 (percent)

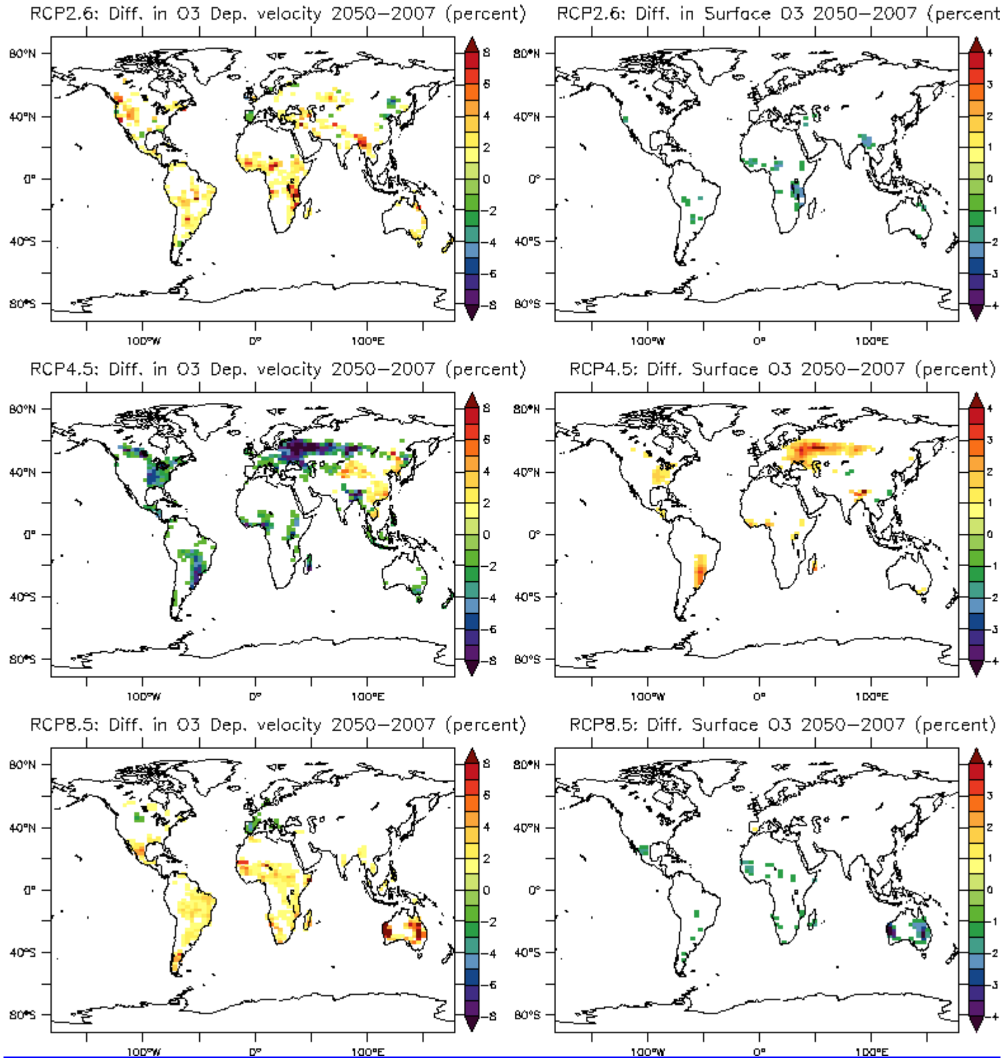
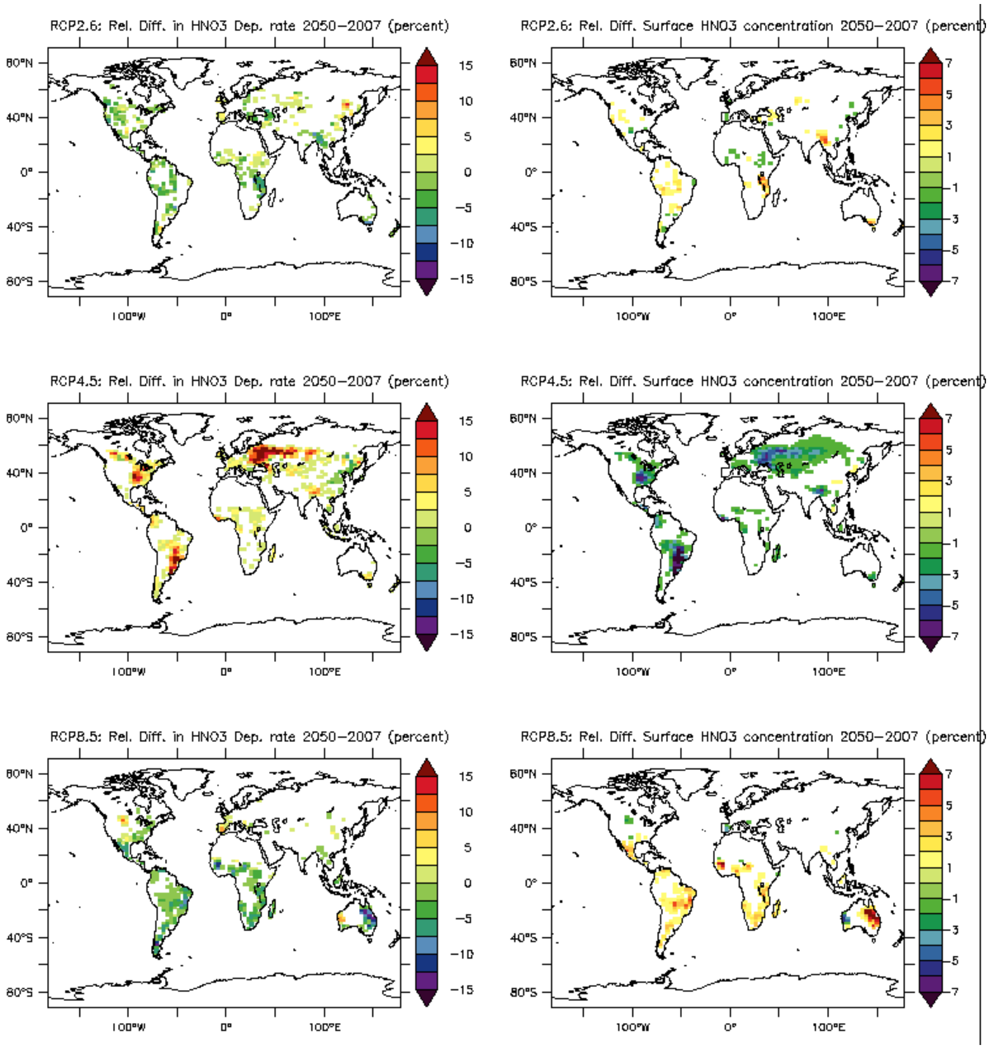


Figure ~~5~~6: Annual mean changes (in relative value %) of ~~Dry~~surface dry deposition velocity for O_3 between present-day and 2050 induced by the different LCC (Left) and related surface ozone concentrations (Right) for the three RCP scenarios. Values in the [-1;+1]% interval are not shown

Mis en forme : Indice



RCP2.6: Rel. Diff. in HNO3 Dep. rate 2050−2007 (percent)

RCP2.6: Rel. Diff. Surface HNO3 concentration 2050−2007 (percent)

RCP4.5: Rel. Diff. in HNO3 Dep. rate 2050−2007 (percent)

RCP4.5: Rel. Diff. Surface HNO3 concentration 2050−2007 (percent)

RCP8.5: Rel. Diff. in HNO3 Dep. rate 2050−2007 (percent)

RCP8.5: Rel. Diff. Surface HNO3 concentration 2050−2007 (percent)

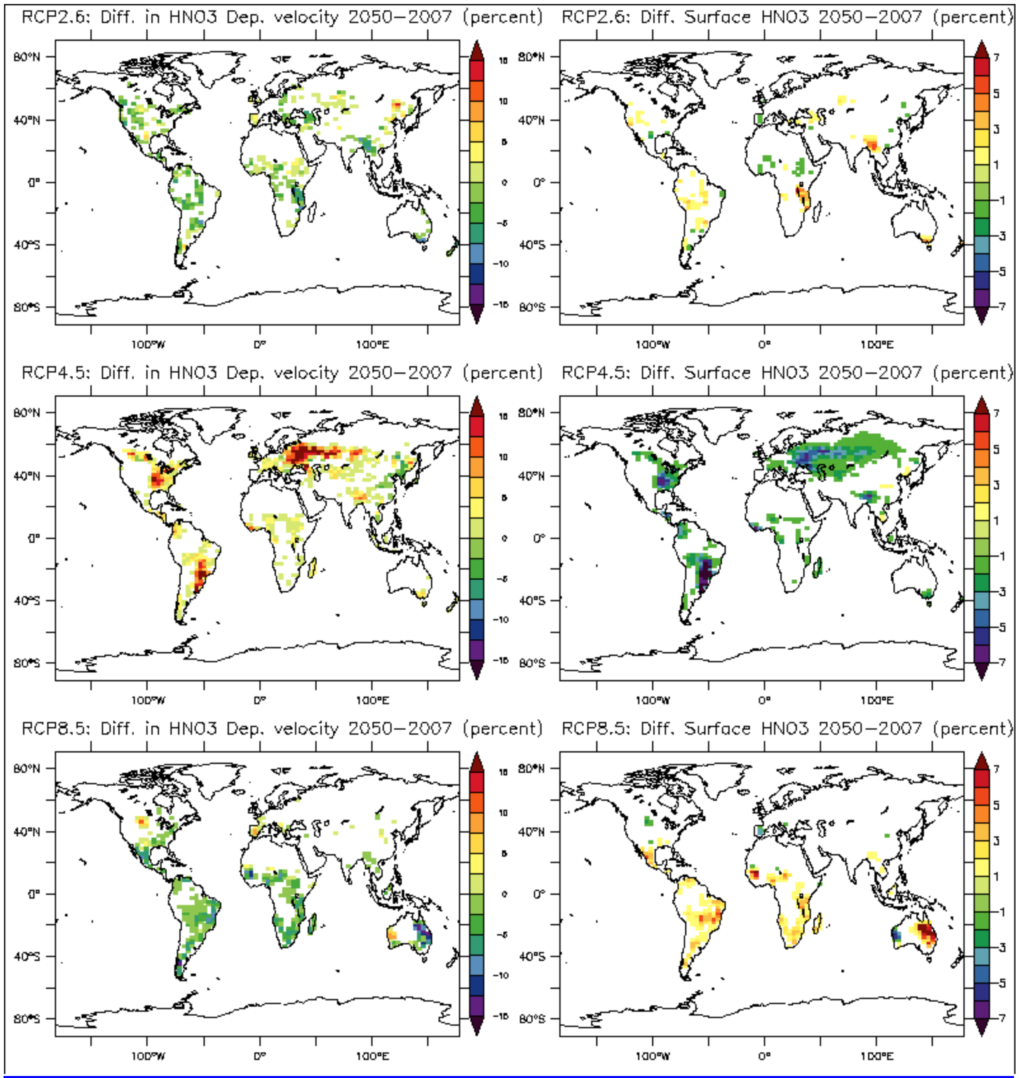
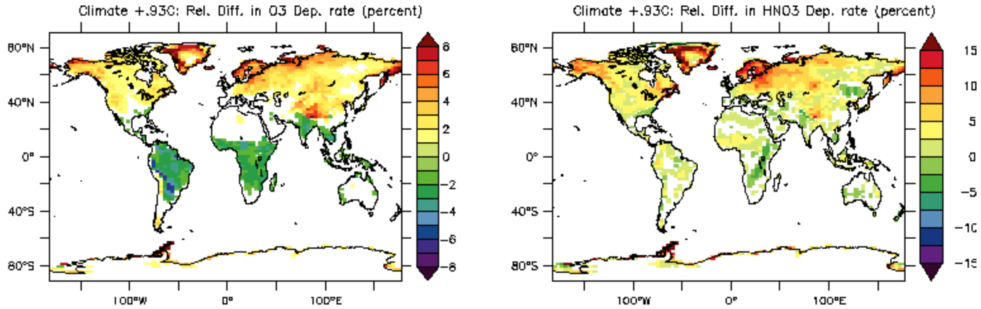


Figure ~~6~~7: Same as Figure ~~5~~6 for HNO_3.

Mis en forme : Indice



Climate +.93C: Rel. Diff. in O3 Dep. rate (percent)

Climate +.93C: Rel. Diff. in HNO3 Dep. rate (percent)

Figure ~~7~~8: Future climate-induced impacts on surface dry deposition velocities (%) considering a 0.93°C increase of global temperature.

Mis en forme : Centré