Anonymous Referee #2

Received and published: 8 September 2010

This paper discusses the specific role of the transport sector (and its split between air, land and sea) on atmospheric chemistry in 2000 and 2050. This is a relevant study that should be published after the authors address the various comments and questions.

Main comment:

The paper lacks clarity in exactly what the authors are discussing. The terminology can be quite vague throughout the paper, making it difficult for the reader to know if the authors are describing the role of the transport sector only or the overall changes in emissions. I would strongly encourage the authors to read their paper carefully and make sure that any discussion of impact is well-defined. What is actually being discussed? Maybe the use of acronyms would help.

While changes in all anthropogenic emissions are considered in the future emission experiments (experiments B to E), only induced changes in the perturbation by the transport sector is discussed. In fact, as explained in p15764, *"it is also of great importance to use realistic 'present' and future emissions by other emission sectors, since the ozone sensitivity to traffic emissions substantially depends on background precursor concentrations".*

To clarify this point, the first sentence of section 4.2 (p15773, L.10) has been corrected as follows:

Initial sentence

In this section, we assess the effect of future changes in traffic emissions on tropospheric ozone

Final sentence

In this section, we describe the impact of the future changes in anthropogenic emissions on the role of the transport sector on the global tropospheric chemistry.

The following sentence has been also added in the conclusion (p15782, line 6):

While changes in all anthropogenic emissions are considered by 2050, only induced changes in the role of the transport sector on the tropospheric chemistry is discussed.

Comments:

Page 15756, line 11: change "amoderate" to "a moderate". Ok

Page 15756, line 12: nothing can be expected, only projected.Ok

Page 15756, line 14: change "adrastic" to " a drastic" Ok

Page 15757, line 16: change "sensitivity" to "sensitive" Ok

Page 15758, line 9: "increasing" over what? This is what I refer to in my main comment. "Increasing" has been changed to "contributing" as follows:

Initial sentence

More recently, Matthes et al. (2007) concluded that the maximum relative impact of road traffic emissions (NOx, CO, NMHC) occurs at northern mid-latitudes in July, increasing the surface and the zonal mean ozone concentrations by up to 16% and 12%, respectively

Final sentence

More recently, Matthes et al. (2007) concluded that the maximum relative impact of road traffic emissions (NOx, CO, NMHC) occurs at northern mid-latitudes in July, contributing by up to 16% and 12% of the surface and zonal mean ozone concentrations, respectively.

Page 15758, line 24: "better" than what? "better" compared to road traffic (as mentioned in the sentence). Changed to "higher"

Page 15760, line 29: change "given" to "specific" Ok

Page 15762, line 23: there is a need for reference to the final QUANTIFY emissions done Borken-Kleefeld et al., 2010 has been added.

Page 15764, line 5: it is unclear if the met. fields are exactly the same in all the perturbations simulations This is explained in the specific section 3.3, which describes the different simulations (p. 15767-15769), as well as in the legend of Table 2 which states: *"The A to C experiments correspond to nudged simulations (2002–2003, ECMWF*

meteorology) and the D to F experiments to 10 yr climatic simulations (GCM mode)".

Also, regardless of the change in stratospheric ozone, strattrop exchange will still be affected by changes in the vertical circulation across the tropopause. We agree. Therefore, the sentence has been modified and completed as follows:

Initial sentence:

This climatology is deliberately kept fixed at present-day values in all simulations in order to isolate the effects of tropospheric chemistry and climate change to changes in the chemical composition of the stratosphere.

Final sentence:

This climatology is deliberately kept fixed at present-day values in all simulations in order to isolate the effects of tropospheric chemistry and climate change to changes in the stratospheric chemistry. Regardless the prescribed stratospheric ozone, the stratosphere to troposphere exchanges are affected by changes in the vertical circulation across the tropopause.

Page 15765, line 8: which "activity"? The whole sentence is about biomass burning activity. We deleted "activity" in the sentence.

Page 15765, line 20: refer to Table 1.Ok Page 15769, line 16: is it only the ozone perturbation that does not affect the climate? How do you separate? In our simulations, none of the changes in the tropospheric chemistry due to changes in emission affects the climate because the climate is fixed from HadGEM1 simulation outputs. This has been clarified as follows:

Initial sentence (Page 15769, lines 18-20):

It must be emphasized here that the ozone perturbation due to traffic emissions is not included in the climate perturbation, i.e., ozone changes do not feedback on climate.

Final sentence:

It must be emphasized here that the present (experiments A to E) and future (experiment F) climates are forced using HadGEM1 simulation outputs, and therefore, that the changes in the tropospheric chemistry simulated in this study do not feedback on climate.

Page 15769, line 26: change "most recent emission data" to "our base inventory". "our base run inventory" is used instead.

Page 15770, line 8: the whole discussion of NOx titration is very resolution dependent and the resolution of LMDz is quite coarse. This has to be discussed here. I would guess that the resolution used here leads to less titration than in the real world.

We agree with this remark. Due to the coarse resolution of the model, the small scale titration effects, such as the one which takes place inside the emission plumes, are not taken into account in this study. These potential effects and the way they can affect our conclusions on the transport-induced ozone are discussed in Cariolle et al. (2010) and Huszar et al. (2010) for aircraft and shipping, respectively. These effects are also summarized in the introduction which has been updated as follows:

p15759 line 5

New text

Huszar et al. (2010) found that the contribution of ship induced surface NOx to the total background reaches 90% over remote ocean and makes 10–30% near coastal regions. They calculated that due to ship emissions, surface ozone increases by up to 4–6 ppbv making 10% contribution to the surface ozone budget.

p15759 line 25

Initial text

.. (Cariolle et al., 2009). This last study, which more especially focused on the assessment of sub-grid effects, also reported that the non-linear chemistry which takes place in the plume dispersion leads to a decrease of the aircraft-induced ozone production by 10 to 15% in the Northern Hemisphere, with the largest effects in the North Atlantic Flight Corridor. **Final text**

.. (Cariolle et al., 2009). This last study and Huszar et al. (2010) study more especially focused on the assessment of plume sub-grid effects. They calculated that the non-linear chemistry which takes place in the plume dispersion leads to a decrease of the aircraft and the shipping-induced ozone production, by 10 to 15 % (free troposphere of northern hemisphere) and by 15 to 30% (surface ozone over the Eastern Atlantic), respectively.

New reference:

Huszar, P., Cariolle, D., Paoli, R., Halenka, T., Belda, M., Schlager, H., Miksovsky, J., and Pisoft, P.: Modeling the regional impact of ship emissions on NO_x and ozone levels over the Eastern Atlantic and Western Europe using ship plume parameterization, Atmos. Chem. Phys., 10, 6645-6660, doi:10.5194/acp-10-6645-2010, 2010.

Page 15770, line 14: define how the tropospheric column is computed.

The thermal tropopause is computed by the general circulation model. This diagnostic is used to compute, during the simulation, the tropospheric ozone column by multiplying in the levels lower than the tropopause, the ozone volume mixing ratio by the density of the column (and then by a conversion factor to have Dobson unit).

Page 15770, line 25: "Land-based emissions" of what?

"Land-based emissions" changed to "road traffic emissions" in the whole document.

Page 15771, line 2: remove "latitude" Ok

Page 15772, line 9: rewrite "corresponding to over" "corresponding to" removed

Page 15775: this whole discussion is quite repetitive. Try shortening. Shortened from 556 words to 388 words, as follows

Initial text (Page 15775-15776): 4.2.2 B1 ACARE mitigation

In order to assess the efficiency of possible emission reduction strategy in view of limiting the future impact of aircraft, the B1 ACARE scenario has been simulated for 2050. It corresponds to B1 2050 emission scenario, but with a reduction of the aircraft emissions due to the implementation of ACARE targets (Table 3). The scenario is characterized by the lowest NO_x global aircraft emissions, i.e., even lower than the 2000 emissions. It can be seen as a more restricting but technically feasible scenario. The ozone perturbation due to aircraft emissions for the B1 2050 ACARE scenario is illustrated and compared to 2000 and B1 2050 perturbations in Fig. 6. A 5% perturbation of aircraft emissions leads to a perturbation of less than 0.10% of the zonal mean ozone background of the upper troposphere of Northern Hemisphere, instead of up to 0.12% and 0.14% for 2000 and B1 2050 emissions, respectively (Fig. 6a). The ozone column

perturbations show that the implementation of the B1 ACARE 2050 targets leads to a decrease of 25% to 36% of the impact of aviation compared to B1 2050 (not shown). The higher relative effect is simulated over the Southern Hemisphere and around the equator. As a consequence, whereas for B1 scenario, the aircraft impact increases over most of the globe from 2000 to 2050 (except over a region including South of US and a Western part of the North-Atlantic ocean), the B1 ACARE scenario leads to an increase in the Southern Hemisphere (by up +30%), but a decrease in the Northern Hemisphere (by down to -30%) compared to 2000 (Fig. 6b). Aviation still remains the major transport contributor to the ozone perturbation in the Northern Hemisphere, by contributing by up to 70% to the transport perturbation on the O3 column (not shown), instead of 85% for the B1 scenario without mitigation (Fig. 2b).

Figure 7a shows the sensitivity of the global ozone burden to each transport mode, for 2000, 2025 and 2050 emissions. The predominant perturbation mode shifts from road to aircraft (A1 scenario) or shipping (B1 scenario) from 2000 to 2050. This shift is due to changes in the respective emission amounts, whereas the ozone production efficiency (O₃ molecule per NO_x molecule emitted) of each transport sector is nohighly modified, despite changes in the background chemical composition (Fig. 7b). In agreement with Hoor et al. (2009), aircraft NO_x emissions have an ozone production efficiency about three times higher than road traffic

and shipping. In fact, aircraft emit directly into the UTLS, where NO_x and ozone have a longer lifetime and accumulate, so leading to larger and more persistent perturbations compared to the Earth's surface, where the exhaust products are more rapidly removed by scavenging and dry deposition (e.g., Hauglustaine et al., 2005; Gauss et al., 2006; Dahlmann et al., 2009). However, as already mentioned, the shipping emissions have the highest net chemical production per NO_x molecule (Fig. 7c). This is due to the fact these emissions largely occur in low polluted environments. Our results are also in accordance with Dahlmann et al. (2009) who found that future changes in ozone production by the transport sector strongly depend on the respective contributions of the three transport modes (because of their different ozone production efficiency), whereas changes in the atmospheric background composition only slightly modify the ozone production efficiency from each source.

Final text

4.2.2 B1 ACARE mitigation

In order to assess the efficiency of possible aircraft emission reduction strategy, the B1 ACARE scenario was simulated for 2050. It corresponds to B1 emission scenario, but with a reduction of the aircraft emissions due to the implementation of ACARE targets (Table 3). It is characterized by low NO_x global aircraft emissions (lower than for 2000), and can be seen as a more restricting but technically feasible scenario. The induced ozone perturbation for this scenario is illustrated and compared to 2000 and B1 2050 perturbations in Figure 6. A 5% perturbation of aircraft emissions leads to a perturbation of less than 0.10% of the zonal mean ozone background of the upper troposphere of Northern Hemisphere, instead of up to 0.12 % and 0.14% for 2000 and B1 2050 emissions, respectively (Figure 6a). The ozone column perturbations show a decrease of 25% to 36% of the impact of aviation compared to B1 2050 (not shown). Whereas for B1 scenario, the aircraft impact increases over most of the globe from 2000 to 2050, the B1 ACARE scenario leads to an increase in the Southern Hemisphere (by up +30%), but a decrease in the northern hemisphere (by down to -30%) (Fig 6b). Nevertheless, aviation would still be the major transport contributor to the ozone perturbation in the Northern Hemisphere in 2050, by contributing by up to 70% to the total transport perturbation on the O₃ column (not shown).

Figure 7a shows the sensitivity of the global ozone burden to each transport mode, for 2000, 2025 and 2050 emissions. The predominant perturbation mode shifts from road to aircraft (A1 scenario) or shipping (B1 scenario) from 2000 to 2050. This shift is due to changes in the respective emission amounts, whereas the ozone production efficiency (O_3 molecule per NO_x molecule emitted) of each transport sector is not highly modified (Figure 7b). In agreement with previous studies (e.g., Hauglustaine et al., 2005; Gauss et al., 2006; Dahlmann et al., 2009, Hoor et al., 2009), aircraft NO_x emissions show an ozone production efficiency about three times higher than road traffic and shipping, because they are emitted directly into the UTLS, where they lead to larger and more persistent perturbations compared to the Earth's surface. The shipping emissions, which largely occur in low polluted environments, have the highest net chemical production per NO_x molecule (Figure 7c).

Page 15779: Figures 9 and 10 are quite noisy. Are the differences shown statistically significant or is it just a display of internal variability and changes in circulation.

As mentioned in the text, "The general patterns of the impact of A1B 2050 transport emissions on the ozone column are very similar in the future (Figure 10c) and present (Figure 10b) climate". However, "slight but significant positive and negative effects, according to the location and the season, on the ozone change. A climate-induced increase of up to +0.6 DU is simulated in January in both northern (North Pacific) and southern (West coast of South-America) hemispheres (Figure 11a)". In this part of the paper dealing with climate effect, the results and figures presented are based on averages performed using 7 years of simulation as described p 15768 line 21-25. This ensures that we analyze the long term climate change and not the inter-annual variability of the circulation.

Page 15781: One needs to split the discussion of Table 4 into more details. As it stands now, the authors are comparing apples and oranges.

Two Tables are now provided (Table 4 and 5) and referred in the text.

Page 15782, line 15: "aviation becomes the main tropospheric ozone perturbation factor". Is that really true?? Or does "perturbation factor" have to be more appropriately qualified?

We mean "the main transport perturbation". The sentence has been modified as follows.

Initial sentence

As a consequence of a strong reduction in road traffic and a moderate (B1 scenario) to high (A1B scenario) increase in ship and aircraft emissions in 2050, a drastic decrease in the impact of road emissions is projected, whereas aviation becomes the main troposheric ozone perturbation factor in the free troposphere, even in the case of a very optimistic mitigation scenario (B1 ACARE).

Final sentence

As a consequence of a strong reduction in road traffic and a moderate (B1 scenario) to high (A1B scenario) increase in ship and aircraft emissions in 2050, a drastic decrease in the impact of road emissions is projected, whereas aviation becomes the major transport perturbation on ozone concentrations in the free troposphere, even in the case of a very optimistic mitigation scenario (B1 ACARE).

Page 15783, line 16: rewrite sentence starting with "Nevertheless". It does not quite make sense as written.

Initial sentence

Nevertheless, similarly to the ozone background, the transport-induced global ozone burden is lowered (-1.6%), whereas positive or negative climate-induced changes are predicted according

to the region, altitude and season.

Final sentence

While the transport-induced global ozone burden is lowered by 1.6% in a future climate compared to the present climate, positive or negative changes are predicted according to the region, altitude and season.