Inclusion of global studies may strengthen the paper

Volker Grewe[1,2] and Mariano Mertens[1]

[1]Deutsches Zentrum für Luft- und Raumfahrt, Institut für Physik der Atmosphäre, Oberpfaffenhofen, Germany
[2]Delft University of Technology, Aerospace Engineering, Section Aircraft Noise and Climate Effects, Delft, the Netherlands

Congratulations to the authors for this well-written comparison of methods related to the evaluation of emission impacts! We think that an inclusion of similar discussions from the chemistry-climate community would even strengthen the paper. Four aspects may be especially of interest:

1. We believe that the tagging method presented in Section 4.1 is identical to tagging approaches used in global model. (e.g. Horowitz and Jacob, 1999; Lelieveld and Dentener, 2000; Meijer et al., 2000; Grewe, 2004; Gromov et al., 2010; Butler et al.,2011; Emmons et al., 2012; Grewe et al., 2012,2017) Is that correct? Can you comment on this?

2. Are the results of your comparison deviating from earlier intercomparisons between the two methods (source apportionment and sensitivity analysis)? Here we refer to Grewe (2013) where the simple differential equation

$$\frac{dx}{dt} = P - x^{\alpha}, \tag{1}$$

   with x the concentration of a chemical species, is used to analyse the differences between the two approaches. Further, we refer to Grewe et al. (2010) where a more complex chemical system (3 species: pseudo NO_y, pseudo VOC and pseudo O_3 and 3 differential equations) is investigated. This system shows the characteristics similar to that of tropospheric ozone chemistry and is hence well suited as a test bed. This system was evaluated with respect to the differences between both methods. Grewe et al. (2010) and Grewe (2013) also nicely show that both methods are equal in a linear regime, which supports the author's statement.

3. We agree that depending on the purpose of the analysis one or the other method is appropriate, as discussed by the authors. However, we miss a clear statement that the combination of both methods give a much better basis for the interpretation of mitigation options.

4. On top of the previous point, a more in depth discussion on the effectiveness and suitability of the discussed methods for the use in political and economic frameworks, which try to provide incentives for mitigation

options might be helpful to understand the implications of using those methods. For example, only the sensitivity method (or also called perturbation method) is suitable to evaluate the impact of a mitigation measure. However, while having said this, deducing from this statement that only the sensitivity measure should be used in any political and economic framework for incentivising measures, might be plainly false. We think that this is an important point which should be raised in the paper. Let's take exemplarily the response of the ozone chemistry on a decrease of NO_x emissions (see Fig. 1). In a saturated chemical regime a decrease of NO_x from e.g. road traffic (Fig. 1a) might not lead to any change in the ozone concentration (Fig. 1b). As NO_x emission decrease, but not the ozone concentration, the net-ozone production (NO3P) per NO_x-molecule (Fig. 1c) increases.

Hence this mitigation measure leads to important change in the chemistry without affecting the ozone concentration. The chemistry might have left the saturation regime because of this first measure. If a second NO_x emission reduction is implemented, this second measure reduces the ozone concentration, since the chemical regime is not anymore saturated and hence takes advantage of the first measure. An ozone reduction is only achieved because the first measure took place. Assessing the two measures by the sensitivity analysis makes the effectiveness prone to the order of implementation. This has been touched in the conclusion (line 802), but might be clarified. On the other hand tagging methods would be more suitable in assessing the effectiveness of mitigation measures, since they are largely (of course not totally) independent of the order of implementation. This is sketched in Fig 1d). The perturbation approach would not see a change of the road traffic impact, as the ozone concentration would not change. The contribution, however, as calculated with a tagging method would be lowered as less NO_x molecules from road traffic are involved in the production of ozone.

A first discussion of these effects were given in Grewe et al. (2012) and will further discussed in a paper which we will submit this summer to ACPD (Mertens et al. 2017). The Fig. 2 is taken from that paper and nicely illustrates the impact of the degree of saturation of a chemical regime on the assessment of mitigation options.

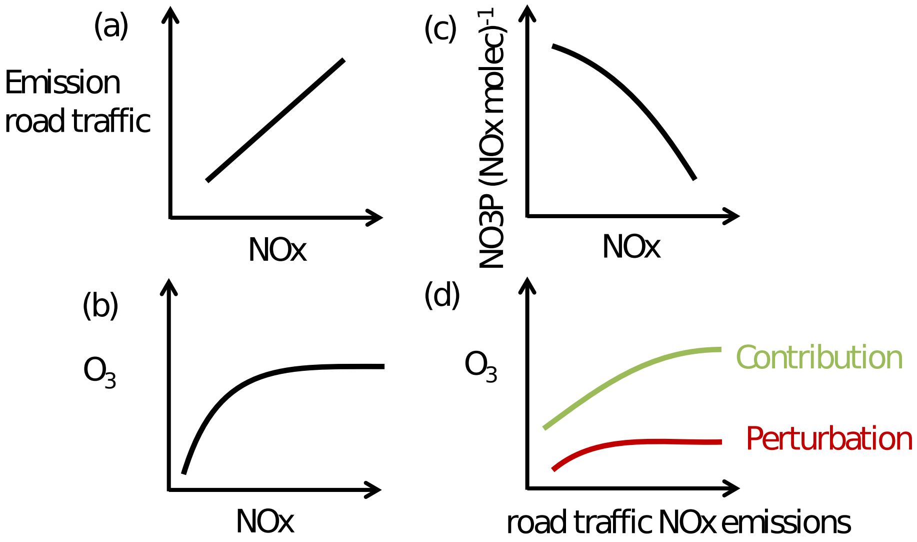

Figure 1: Response of the ozone concentration of an emission reduction (details are given in the text).

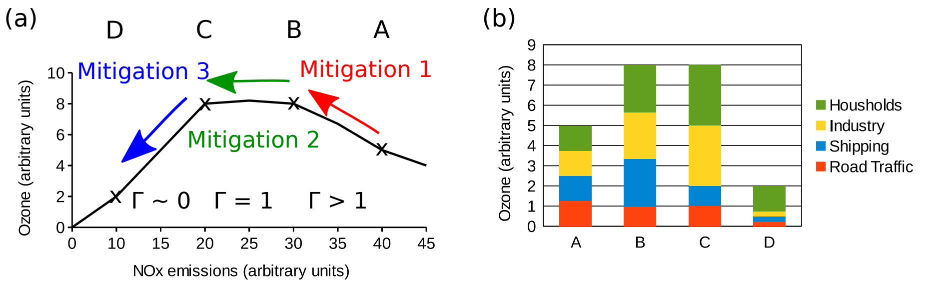

Figure 2: Sketch showing the dependence of the successes of a mitigation measure on the history of previous mitigation measures. Taken from Mertens et al. 2017

References:

Butler, T. M., Lawrence, M. G., Taraborrelli, D., and Lelieveld, J.: Multi-day ozone production potential of volatile organic compounds calculated with a tagging approach, Atmos. Environm. 45, 4082-4090, 10.1016/j.atmosenv.2011.03.040, 2012.

Emmons, L. K., Hess, P. G., Lamarque, J.-F., and Pfister, G. G.: Tagged ozone mechanism for MOZART-4, CAM-chem and other chemical transport models, Geosci. Model Dev., 5, 1531-1542, 10.5194/gmd-5-1531-2012, 2012.

Grewe, V.: Technical Note: A diagnostic for ozone contributions of various NOx emissions in multi-decadal chemistryclimate model simulations, Atmos. Chem. Phys. 4, 729-736, 10.5194/acp-4-729-2004, 2004.

Grewe, V., Tsati, E., and Hoor, P.: On the attribution of contributions of atmospheric trace gases to emissions in atmospheric model applications, Geosci. Model Dev. 3, 487-499, 10.5194/gmd-3-487-2010, 2010.

Grewe, V., Dahlmann, K., Matthes, S., and Steinbrecht, W.: Attributing ozone to NOx emissions: Implications for climate mitigation measures, Atmos. Environ. 59, 102-107, 2012.

Grewe, V.: A generalized tagging method, Geosci. Model Dev., 6, 247-253, 10.5194/gmd-6-247-2013, 2013b.

Grewe, V., Tsati, E., Mertens, M., Frömming, C., and Jöckel, P., Contribution of emissions to concentrations: The TAGGING 1.0 submodel based on the Modular Earth Submodel System (MESSy 2.52), Geosci. Model Dev. 10, 2615-2633, 10.5194/gmd-2016-298, 2017.

Gromov, S., Jöckel, P., Sander, R., and Brenninkmeijer, C. A. M.: A kinetic chemistry tagging technique and its application to modelling the stable isotopic composition of atmospheric trace gases, Geosci. Model Dev., 3, 337-364, 10.5194/gmd-3-337-2010, 2010.

Horowitz, L. and Jacob, D., Global impact of fossil fuel combustion on atmospheric NOx , J. Geophys. Res., 104, 23823-23840, 10.1029/1999JD900205, 1999.

Lelieveld, J. and Dentener, F. J.: What conrols tropospheric chemistry, J. Geophys. Res., 105, 3531-3551, 2000.

Meijer, E., van Velthoven, P., Thompson, A., Pfister, L., Schlager, H., Schulte, P., and Kelder, H.: Model calculations of the impact of NOx from air traffic, lightning, and surface emissions, compared with measurements, J. Geophys. Res., 105, 3833-3850, 2000.

Mertens, M., Grewe, V., Rieger, V.S., Jöckel, P., The contribution of road traffic and shipping emissions to tropospheric ozone revisited, in preparation to ACPD, August 2017.