Atmospheric Environment 59 (2012) 102–107

Contents lists available at SciVerse ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

# Attributing ozone to $NO_x$ emissions: Implications for climate mitigation measures

Volker Grewe [a,*], Katrin Dahlmann [a], Sigrun Matthes [a], Wolfgang Steinbrecht [b]

[a] Deutsches Zentrum für Luft- und Raumfahrt, Institut für Physik der Atmosphäre, Oberpfaffenhofen, Germany
[b] Meteorologisches Observatorium Hohenpeienberg, Deutscher Wetterdienst, Hohenpeienberg, Germany

## HIGHLIGHTS

- We calculate contribution of $NO_x$ emissions to ozone concentrations.
- The perturbation method largely underestimates the road traffic contribution to ozone.
- Combination of perturbation and tagging methods improves understanding.
- We propose to use the tagging method to evaluate mitigation measures.

## ARTICLE INFO

*Article history:*
Received 19 February 2012
Received in revised form
26 April 2012
Accepted 1 May 2012

*Keywords:*
$NO_x$ emissions
Climate change mitigation
Ozone chemistry
Tagging

## ABSTRACT

Emissions of nitrogen oxides ($NO_x$) lead to formation of ozone, which is an important greenhouse gas. Despite its relevance, little emphasis was previously given on verifying approaches to calculate contributions of individual emissions to ozone and hence to climate change. Basically two methods (perturbation method and tagging method) were used in the past. We demonstrate that both methods are valid and have their area of application, but only tagging calculates contributions of emissions to concentrations, whereas the perturbation method identifies changes in the ozone concentrations due to emission changes. Our results show that the contribution of road traffic emissions to climate change is underestimated by a factor of 5 in the perturbation method. This is caused by non-linear compensating effects from other emission sectors, which are concealed in the perturbation method but disclosed with tagging. Consequently, the effectiveness of mitigation measures for individual sectors (i.e. concentrating on road traffic induced ozone) is only correctly expressed by the tagging method. The perturbation method provides accurately the total impact (i.e. total ozone) of a mitigation measure. However, current approaches, which evaluate the effectiveness of a mitigation measure based on the perturbation approach, do not reflect changes in the chemical state of the atmosphere (i.e. ozone production rates). These largely affect the effectiveness of subsequent measures and hence make the evaluation of the effectiveness of two measures dependent on their chronology of application. We show that also in this regard, the tagging method is better suited to evaluate the effectiveness of a mitigation measure than the perturbation method.



## 1. Introduction

Climate change is a challenge to society. Identification of promising mitigation measures requires a good understanding of the individual contributions of anthropogenic emissions, e.g. from aviation, shipping and road traffic. These contributions were recently estimated in reviews (Lee et al., 2010; Eyring et al., 2010; Uherek et al., 2010). The studies used state-of-the art numerical models, and compared a base case with "all emissions" to a perturbation case with "changed emissions". However, this "perturbation method" may not be adequate for emissions like nitrogen oxides ($NO_x$) which have highly non-linear effects on ozone chemistry. Instead, a "tagging method" (Wang et al., 2009; Grewe et al., 2010) was suggested for the calculation of these contributions. Tagging implements an accounting system that follows the fate of emitted species and keeps track of their chemical reaction pathways. Thereby, the contribution of an emission to ozone is inherently defined as the fraction of current ozone due to the particular source.

The atmospheric concentration of $NO_x$ controls the non-linear character of atmospheric ozone chemistry (Liu et al., 1987; Lin et al., 1988; Grooß et al., 1998): At low $NO_x$ concentrations, ozone

* Corresponding author.
*E-mail address:* volker.grewe@dlr.de (V. Grewe).

1352-2310/$ – see front matter © 2012 Elsevier Ltd. All rights reserved.
doi:10.1016/j.atmosenv.2012.05.002

production is limited by the availability of $NO_x$ molecules. At high $NO_x$ concentrations, other compounds limit ozone production, e.g. hydrogen oxide radicals ($HO_x$) (Lin et al., 1988) (see also Fig. 1a). Additional ozone destruction by reactions with $NO_x$ becomes increasingly important at the highest $NO_x$ concentrations. Eventually it even leads to negative net ozone production rates (Dahlmann et al., 2011, e.g.). Ozone is an important greenhouse gas. Hence $NO_x$ related ozone changes have an impact on climate that is more complex than climate effects of $CO_2$ increases. Significant increase in surface ozone has been observed at northern hemisphere mid-latitudes in the 1970s and 1980s (Oltmans et al., 2006). More recently, at some locations ozone concentrations have leveled off or even declined slightly, whereas others still show an increase (Oltmans et al., 2006; Parrish et al., 2009). Much of the ozone trends can be attributed to precursor emissions. Other contributions come from the decline of stratospheric ozone and from changes in stratospheric ozone intrusions (Grewe, 2007). $NO_x$ precursor emissions from road traffic saw a large increase in the 1970s and 1980s, but later decreased due to the wide-spread implementation of catalytic converters (Uherek et al., 2010). Estimates of the contribution of road traffic emissions to northern hemispheric ozone are based on numerical simulations with atmosphere-chemistry models. They range from 2% to 12% for summer (Uherek et al., 2010). However, usually these numbers are obtained by the "perturbation method". In Section 4, we show that this approach only gives the sensitivity for adding more $NO_x$, but not the actual contribution from a single sector. Note that these two numbers would agree in a linear chemical system, but they differ substantially in non-linear chemical regimes (Grewe et al., 2010).

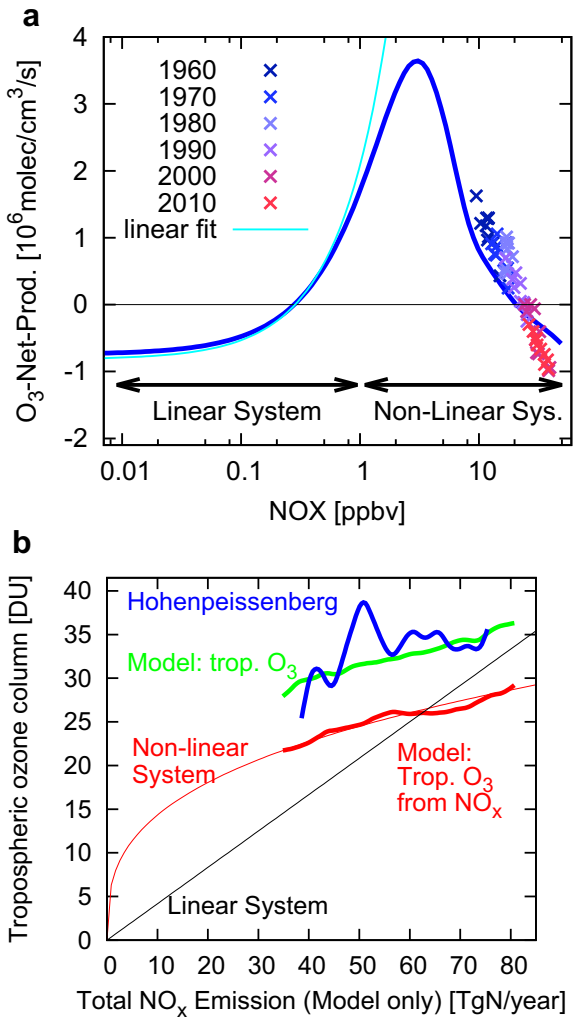


**Fig. 1.** Non-linearities in tropospheric ozone. a) Net ozone production rates at 50°N in Europe for summer conditions. The solid blue line shows results based on box model simulations, which were constrained by values from the global chemistry climate simulation. The crosses indicate the results from the global simulation and are color-coded with respect to time from 1960s to 2010s. Note that the presentation is logarithmic. The light blue line shows a linear fit, which appears in the logarithmic scale as an exponential function and gives an indication, where $NO_x$-ozone relation is linear. b) Relation between simulated global tropospheric ozone columns and total $NO_x$ emissions (green line), with and without contributions from stratospheric ozone (red line). A fit (square root function) is added (thin red line) and a linear function (black line) is added indicating the non-linear character of the $NO_x$-ozone relationship. Measurement data from Hohenpeissenberg (ground to 11 km) are added (blue line) by allocating the total $NO_x$ emissions prescribed in the model to the time of the measured data. All data are smoothed and based on annual means. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

It has to be stressed that both methods were previously applied in many investigations. The tagging method to identify contributions from individual sources (Horowitz and Jacob, 1999; Leliveld and Dentener, 2000; Meijer et al., 2000; Grewe, 2004) and perturbation approach to investigate atmospheric sensitivities to individual sources (Horowitz et al., 1998; Stevenson et al., 2006). Both methods are useful and target different objectives, but they are not interchangeable (Wang et al., 2009; Grewe et al., 2010). In this study we will show first that the combination of both methods lead to a better analysis of mitigation measures and second that an evaluation of a mitigation measure, which purely concentrates on the total effect on ozone, insufficiently describes the effect of this measure.

We briefly introduce the applied methodology, the models and the contribution calculation in the following Section. The key difference in the perturbation method and the tagging method arises from non-linearities in the chemical regime, which are presented in more detail in Section 3. The results for the contribution calculation are shown in Section 4 and the implication for the evaluation of mitigation measures is analyzed in Section 5.

## 2. Methodology

### 2.1. Model descriptions

We applied a state-of-the art climate-chemistry model (E39C) (Dameris et al., 2005) for the troposphere and stratosphere to simulate atmospheric composition changes from 1960 to 2019 in an ensemble approach. The model simulations were previously validated and investigated with respect to, e.g. stratospheric ozone depletion (Dameris et al., 2005), lightning (Grewe, 2009), and the chemistry and climate of tropospheric ozone change (Gauss et al., 2006; Grewe, 2007; Dahlmann et al., 2011). The model includes a specific emission-ozone tagging method (Grewe, 2004). This is an accounting system tracking emitted $NO_x$ compounds during transport and chemical processing. It allows the separation of individual source contributions to ozone concentration. Tags are attached to species when they are emitted and passed on to other species in chemical reactions. Two chemical systems are solved: First, atmospheric chemistry, second, the distribution of tags among the chemical species. The tags follow the atmospheric chemical reactions of the first step (Grewe, 2004; Grewe et al., 2010). That means that the differential equations, which describe the chemistry, are solved as in any other atmosphere-chemistry model. This provides production and loss terms for every species. These are employed in the second step to solve differential equations for the contributions (tags) of the individual sources. To every individual emission category new tracers are introduced for $NO_y$

and $O_3$, which are transported, experience the respective emissions, chemical production rates and loss rates. For example the ozone production term for road traffic is the ozone production rate multiplied by the fraction of road traffic $NO_x$ to total $NO_x$. A comparison between the tagging method and the perturbation method is given in Section 2.2. The adjusted radiative forcing calculation (Dahlmann et al., 2011) was performed with the E39 climate model, separately for each tagged ozone field taking into account the seasonal cycle. For Fig. 1a, simulations were performed with the box model Caaba/MECCA (Sander et al., 2011). The methodology follows the approach described in Ehhalt and Rohrer (1994) and Grooß et al. (1998): Longer-lived species and meteorological parameters (e.g. $CH_4$, $N_2O$, CO, $HNO_3$, $HNO_4$, $H_2O$, and temperature) were constrained by values taken from the chemistry climate model simulations for 50°N Europe in July. Short-lived species (e.g. OH and $HO_2$) were allowed to adapt to the situation.

### 2.2. Contribution calculation: the difference between the tagging and perturbation method

The tagging method is an accounting system, which follows the fate of an emission within the chemical system. The perturbation method derives the change in the atmospheric composition caused by a change in an emission by calculating the difference between 2 respective simulations. Both methods are described in detail in literature (Grewe et al., 2010). Here we illustrate the difference between both methods for a practical case (Fig. 2), whereas Grewe et al. (2010) have shown the difference in a theoretical framework. The perturbation method uses a local linearization of the relation between $NO_x$ emission and ozone concentration. This linearization is calculated on the basis of two simulations resulting in a slope triangle, which approximates the derivative. The tagging method follows reaction pathways and traces the fate of an emission within a single simulation (bars on the right). The $y$-intercept of the slope triangle (blue line), calculated with the perturbation method, represents the absolute error of the perturbation method ($\varepsilon_\beta$), i.e. the residuum of the contribution calculation, when it is calculated for every single sector and summed up. In order to reduce computing time we have assumed that the sensitivity of road traffic emission to the ozone concentration is representing the sensitivity of all other emissions. There are deviations to this assumption. However, these have to be small, since the overall sensitivity is given by the derivative to the $NO_x$ emission – ozone curve (Figs. 1b and 2), which is obviously close to that from road traffic. The tagging method includes no residuum, since all partial slopes, independent from their order, have no $y$-intercept.

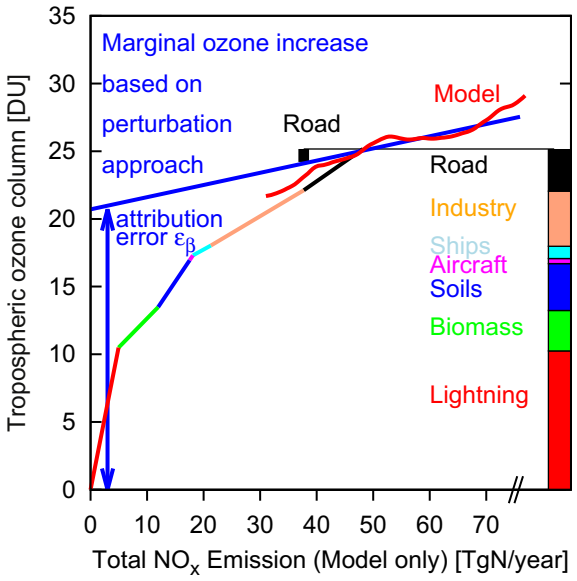


**Fig. 2.** Contribution of individual sectors to tropospheric ozone. As Fig. 1b, with additional information. Simulated tropospheric ozone [DU] from tropospheric photochemistry (red line), identical to that in Fig. 1b. Boxes give the contribution from different sources, calculated with the tagging method for the year 1990. The contribution calculation with the perturbation method is added as an additional black box, which is based on a calculation of a slope triangle, where the slope (blue line) is given by the marginal ozone change for the year 1990, estimated by an additional simulation, which excludes road traffic $NO_x$ emissions. The blue arrow indicates the absolute error $\varepsilon_\beta$ (Grewe et al., 2010), i.e. if all ozone contributions from all individual sources are determined by the perturbation method, always based on a base case simulation and a perturbed simulation. Hence, $\varepsilon_\beta$ is the amount of ozone, which is erroneously not accounted for by the perturbation method. For simplification, the sensitivity of road traffic emissions to ozone is applied to all source categories, which is justified, since the derivative fits visually well. The respective slopes from the tagging method are shown in colors, road traffic in black. Individual emissions have different ozone production efficiencies, indicated by the different partial slopes (color). The order of the individual contributions is arbitrary and the slopes are independent from this order. The perturbation methodology requires two simulations: A base case, here the year 1990, and a perturbation simulation with a change in the emission strength by a factor of $\alpha$. Here we use $\alpha = -100\%$ for road traffic emissions. The difference in both simulations is used to determine the marginal ozone increase per emitted $NO_x$, i.e. mathematically an estimate for the derivative. This is shown by the blue line. It amounts to 0.09 DU of ozone per emission of $NO_x$ [TgN/year], or in other word 0.6 DU for the whole road traffic emission (black box to the left of the intersection). In contrast, the black line and box to the right of the intersection gives the contribution of road traffic emissions on ozone, calculated with the tagging method. This indicates an ozone increase by 0.6 DU per emission of $NO_x$ [TgN/year]. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

## 3. Non-linearity of the chemical regime

We have used comprehensive climate-chemistry ensemble simulations, covering the time span from 1960 to 2019 (Dahlmann et al., 2011; Dameris et al., 2005, 2006; Grewe, 2007). Our model includes a tagging method for all nitrogen sources and their contributions to ozone (Grewe, 2004). An analysis of the net ozone production rates with a box model (Section 2.1) constrained by the global model shows the non-linear character of tropospheric ozone chemistry (Fig. 1a). We see a more or less linear increase of net ozone production rates for low $NO_x$ (Note the logarithmic presentation, which turns a linear fit into an exponential function, light blue line), then saturation, and finally a decrease of net ozone production for high $NO_x$ concentrations. This latter regime is found at northern hemisphere continental sites (Fig. 1a, crosses). Ozone production is clearly not a linear function of $NO_x$ levels. As mentioned, at northern hemispheric continental sites the chemical regime is already far beyond linear. For other locations, the regime is closer to linear, e.g. at 250 hPa over Europe, or in the lower troposphere over Africa (Dahlmann et al., 2011). Transport, mixing, and chemistry act together with local emissions to produce the evolution of global tropospheric ozone columns shown in Fig. 1a.

Globally, increasing $NO_x$ emissions in the period 1960 to 2019 result in an increase in simulated tropospheric ozone (Fig. 1b, green line), which is also apparent from observations, e.g. at Hohenpeissenberg (Fig. 1b, blue line). The red line in Fig. 1 shows only that part of tropospheric ozone that originates from tropospheric photochemistry. The stratospheric contribution is subtracted, (Note that this already requires some form of tagging). This tropospheric ozone vs. $NO_x$ emission curve also gives a clear indication of the saturation in ozone production rates. A 10 TgN/year increase in $NO_x$ emissions from 55 to 65 TgN/year gives little or no increase in ozone (red line). At lower emission levels, however, e.g. from 10 to 20 TgN/year, the same 10 TgN/year increase results in substantial ozone increase. This saturation effect is important. It leads to important differences from a linear chemistry, which would follow e.g. the black line in Fig. 1b. Ultimately, this deviation from linearity

results in a difference between the contributions assigned to a specific emission sector, like traffic, by the "perturbation method" and by the "tagging method" (Grewe et al., 2010).

## 4. Contribution of road traffic emissions

We have applied both methods (tagging and perturbation) in numerical simulations with our climate-chemistry model. Results for 1990 are shown in Fig. 3. Both methods use the exact same simulations: A base case with 1990 emissions, and a "zero road" simulation, where all emissions due to road traffic have been turned off. When using the perturbation method, the difference of the tropospheric ozone columns between the two simulations gives the net ozone column added due to road traffic emissions. There is no additional information. This is symbolized by the two uniform columns on the left of Fig. 3.

The tagging method uses the same simulations, and there is also the same ozone change between the two simulations. However, the additional tagging keeps track of the "chemical fate" of emissions from the different sectors. It allows a true attribution of parts of the ozone columns to different sectors. Only with the tagging method, it is possible to show the individual partial columns in the right part of Fig. 3a. When comparing the "zero road" case with the base case, the overall ozone change is small - the same 0.6 DU as reported by the perturbation method. However, while the 3.5 DU partial ozone column tagged to road traffic vanishes in the "zero road" simulation, all the other partial columns, e.g. those tagged to industry or lightning, have increased by 2.5 DU (see also bottom part of Fig. 3).

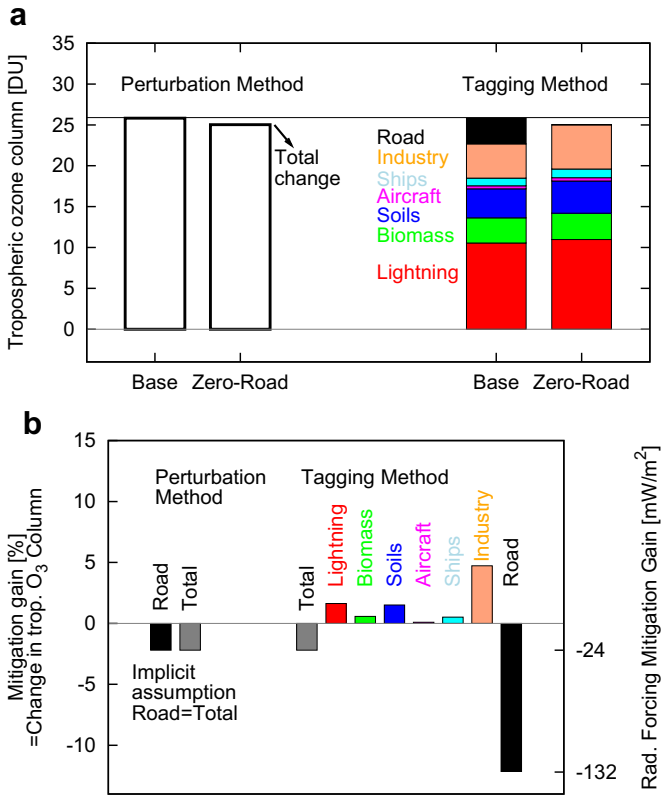


**Fig. 3.** Contribution from different sectors: Illustration of the difference between the perturbation method and the tagging method. a) Tropospheric ozone columns [DU] from $NO_x$ sources for the base case simulation for 1990 (first column) and the respective results from a simulation excluding road traffic emissions (second column). The total change, i.e. the difference in both simulations is attributed to road traffic by the perturbation method (left). The tagging method (right) additionally allows to calculate the contributions/partial columns from the individual emission sectors (colors). The simulations for both methods are identical, but the tagging method provides much more detailed information. b) Relative changes in tropospheric ozone [%, left axis] and radiative forcing [mW $m^{-2}$, right axis] between the two simulations. As above the tagging method (right) provides more detailed information than the perturbation method (left). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

This compensating increase in the other sectors is due to the non-linearities in $NO_x$ chemistry. When road traffic emissions are turned off, the total ozone production decreases slightly. This leads to a little less ozone, but ozone production per $NO_x$ molecule (=production efficiency) does increase, which is in agreement with earlier findings (Lin et al., 1988; Ehhalt and Rohrer, 1994; Grooß et al., 1998). This 2.5 DU increase due to higher production efficiency largely offsets the substantial 3.1 DU from road traffic emissions. Overall, the ozone change amounts to 0.6 DU only – the same small value that the perturbation method "falsely" attributes to road traffic alone. Therefore the perturbation method is well suited to estimate the total ozone change of an alternative emission scenario, but it lacks the possibility of interpreting the results, since compensating effects may conceal a more detailed interpretation.

In terms of a climate metric, e.g. radiative forcing (RF), an indicator for anthropogenic induced climate change, turning off road traffic shows a similar behavior: The RF attributed to road traffic by our tagging method is 132 mW $m^{-2}$. For zero road traffic emissions this would be off-set by 108 mW $m^{-2}$ due to increased ozone production in the other sectors. The overall total mitigation gain would only be 24 mW $m^{-2}$ – the same number as reported by the perturbation method, but roughly 5 times smaller than the 132 mW $m^{-2}$ attributed to road traffic by the tagging method. This large number might be offset by an indirect effect, i.e. a reduction of the natural background ozone by road traffic $NO_x$ emissions, which might be in the order of 10%, taking the low pre-industrial ozone background into account.

## 5. Chronology of mitigation measures

Current emission trading systems consider long-lived trace gases, "Kyoto gases" like carbon dioxide, nitrous oxides and methane. It is widely accepted that emissions of short-lived gases, like $NO_x$ also have considerable impact on climate change (Derwent et al., 2001; Wild et al., 2001; Uherek et al., 2010; Lee et al., 2010; Eyring et al., 2010). Mitigation measures aiming at short-lived effects can reduce the rate of climate change, whereas in order to avoid irreversible climate change, mitigation measures should focus on $CO_2$ (Solomon et al., 2009).

Here, we propose to evaluate mitigation measures by applying the tagging method rather than the perturbation method. We do this because the chosen methodology can have a large effect on the evaluation of mitigation strategies. In the previous section we have already discussed a simplified mitigation scenario for road traffic, where no nitrogen oxides are emitted at all ("zero $NO_x$"). This might be achieved, e.g. through a rigorous implementation of very effective catalytic converters. The perturbation method would give a total ozone change by only 0.6 DU (see Section 4). The wrong conclusion might be that 0.6 DU are caused by road traffic and mitigation is not worthwhile. The tagging method, however, shows that indeed a much larger tropospheric ozone column, 3.1 DU, is tied to road traffic emission. When seen in the context of emissions and mitigation for other sectors, this larger number may have substantial impact on the evaluation of mitigation scenarios. By showing these larger numbers, the tagging method puts a better focus on individual sectors. Measures aiming at reducing the climate impact from a single sector e.g. road traffic can now be evaluated with more detail. It becomes possible to isolate the contribution changes from one sector and to separate them from

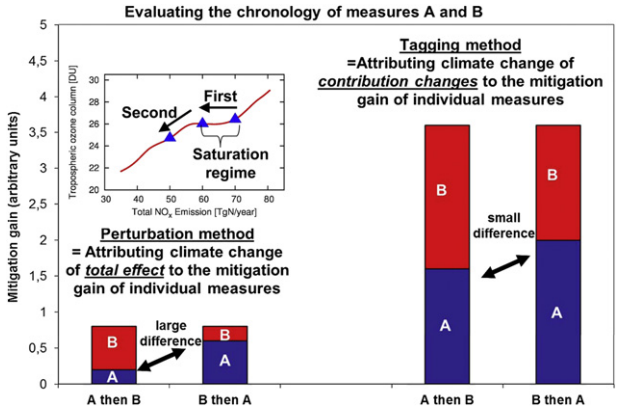


**Fig. 4.** Comparison of the climate change mitigation gain of two arbitrary measures "A" and "B", evaluated with the perturbation method (left) and tagging method (right) for two scenarios in which the order of the two measures changes. Inlay similar to Fig. 1b, but here to show the effect of two mitigation measures. Mitigations "A" and "B" would reduce different sectors by the same amount of $NO_x$ emissions. However, they could be applied in different order: "A" first, then "B", or "B" first, then "A". The sequence could have a substantial effect on the evaluation of each mitigation gain. Based on the perturbation method, only the total change can be considered. Therefore, the first mitigation measure would always have a small impact; the second mitigation would have a larger impact. Using the tagging method, the overall contribution and the net change for each sector can be considered in the evaluation and the temporal order becomes less important. A more balanced evaluation of mitigation measures "A" and "B" becomes possible.

compensating effects from other sectors. Interference between sectors becomes more visible and can be accounted for. This is shown exemplarily in Fig. 4: The base situation is characterized by a saturation (see inlay): any change in $NO_x$ emissions lead to minor changes in ozone. Only after larger $NO_x$ emission reductions the ozone concentration drops significantly due to non-linearities in chemistry, i.e. higher net ozone production rates for lower $NO_x$ concentrations (Section 3). We consider two arbitrary measures "A" and "B", which reduce $NO_x$ emissions. Because of the saturation effect, always the first measure will lead to almost no change in ozone, whereas the second successfully reduces the ozone concentration. Applying the perturbation method will inhibit any measures, since either measure "A" and "B" will be evaluated as ineffective (left part). On the other hand the tagging evaluates either measure as effective independently from the time they are taken into action. The relative difference in the mitigation gain for e.g. measure "A" if becoming effective first or second is small.

In any case the first mitigation measure changes the sensitivity of ozone to $NO_x$ emissions. This effect is important for the effectiveness of any subsequent mitigation measure, but it is not covered by either evaluation approach, respectively. The perturbation method calculates marginal changes for respective sectors, and the tagging method calculates average changes for the respective sectors. Hence the tagging method is less prone to changes in the atmospheric state compared to the perturbation method, which makes it better suited for an evaluation of mitigation measures.

This shows that the evaluation of mitigation measures with the tagging method becomes largely independent of their implementation sequence (Fig. 4), which is not the case for the perturbation method. Overall, the use of the tagging method makes the evaluation of mitigation measures more robust. This should help in our efforts to mitigate anthropogenic climate change.

## 6. Conclusion

We have presented 2 methodologies (perturbation method and tagging), which are widely used to calculate the contribution of individual emission sources to atmospheric composition and applied them to calculate the contribution of road traffic emissions to tropospheric ozone concentrations. The results, i.e. the global ozone mass and radiative forcing caused by road traffic emissions differ by a factor of 5. Tagging can demonstrate that the contribution of road traffic to ozone levels is substantially underestimated by the standard "perturbation method", since gains from $NO_x$ reductions in one sector are partly compensated by more efficient ozone formation from $NO_x$ still emitted in the other sectors. Only the tagging method allows to separate mitigation gains for any single emission sector from non-linear changes induced from remaining emissions in other sectors. Any global post-Kyoto emission trading system that includes short-lived species like $NO_x$ needs to account for these non-linear interactions.

## Acknowledgments

This work was supported by the European project REACT4C and the DLR-project VEU.

## References

Dahlmann, K., Grewe, V., Ponater, M., Matthes, S., 2011. Quantifying the contributions of individual $NO_x$ sources to the trend in ozone radiative forcing. Atmos. Environ. 45 (17), 2860–2868. doi:10.1016/j.atmosenv.2011.02.071.

Dameris, M., Grewe, V., Ponater, M., Deckert, R., Eyring, V., Mager, F., Matthes, S., Schnadt, C., Stenke, A., Steil, B., Brühl, C., Giorgetta, M., 2005. Long-term changes and variability in a transient simulation with a chemistry-climate model employing realistic forcing. Atmos. Chem. Phys. 5, 2121–2145.

Dameris, M., Matthes, S., Deckert, R., Grewe, V., Ponater, M., 2006. Solar cycle effect delays onset of ozone recovery. Geophys. Res. Lett. 33, L03806. doi:10.1029/2005GL024741.

Derwent, R.G., Collins, W.J., Johnson, C.E., Stevenson, D.S., 2001. Transient behaviour of tropospheric ozone precursors in a global 3-D CTM and their indirect greenhouse effects. Clim. Change 49, 463–487.

Ehhalt, D.H., Rohrer, F., 1994. The impact of commercial aircraft on tropospheric ozone. In: Proc. of the 7th BOC Prietly Conference, Lewisburg, Pennsylvania, USA.

Eyring, V., Isaksen, I.S.A., Berntsen, T., Collins, W.J., Corbett, J.J., Endresen, O., Grainger, R.G., Moldanova, J., Schlager, H., Stevenson, D.S., 2010. Transport impacts on atmosphere and climate: shipping. Atmos. Environ. 44, 4735–4771. doi:10.1016/j.atmosenv.2009.04.059.

Gauss, M., Myhre, G., Isaksen, I.S.A., Grewe, V., Pitari, G., Wild, O., Collins, W.J., Dentener, F.J., Ellingsen, K., Gohar, L.K., Hauglustaine, D.A., Iachetti, D., Lamarque, J.F., Mancini, E., Mickley, L.J., Prather, M.J., Pyle, J.A., Sanderson, M.G., Shine, K.P., Stevenson, D.S., Sudo, K., Szopa, S., Zeng, G., 2006. Radiative forcing since preindustrial times due to ozone change in the troposphere and the lower stratosphere. Atmos. Chem. Phys. 6, 575–599.

Grewe, V., 2004. Technical note: a diagnostic for ozone contributions of various $NO_x$ emissions in multi-decadal chemistry-climate model simulations. Atmos. Chem. Phys. 4, 327–342.

Grewe, V., 2007. Impact of climate variability on tropospheric ozone. Sci. Total Environ. 374, 167–181.

Grewe, V., 2009. Impact of lightning on air chemistry and climate. In: Betz, Hans Dieter, Schumann, Ulrich, Laroche, Pierre (Eds.), Lightning: Principles, Instruments and Applications Review of Modern Lightning Research. Springer Verlag, pp. 524–551.

Grewe, V., Tsati, E., Hoor, P., 2010. On the attribution of contributions of atmospheric trace gases to emissions in atmospheric model applications. Geosci. Model Dev. 3, 487–499. doi:10.5194/gmdd-3-819-2010.

Grooß, J.-U., Brühl, C., Peter, T., 1998. Impact of aircraft emissions on tropospheric and stratospheric ozone. Part I: chemistry and 2-D model results. Atmos. Environ. 32, 3152–3184.

Horowitz, L.W., Liang, J., Gardner, G.M., Jacob, D.J., 1998. Export of reactive nitrogen from North America during summertime: sensitivity to hydrocarbon chemistry. J. Geophys. Res. 103, D11. doi:10.1029/97JD03142.

Horowitz, L., Jacob, D., 1999. Global impact of fossil fuel combustion on atmospheric $NO_x$. J. Geophys. Res. 104 (D19), 23823–23840. doi:10.1029/1999JD900205.

Lee, D.S., Pitari, G., Grewe, V., Gierens, K., Penner, J.E., Petzold, A., Prather, M.J., Schumann, U., Bais, A., Berntsen, T., Iachetti, D., Lim, L.L., Sausen, R., 2010. Transport impacts on atmosphere and climate: aviation. Atmos. Environ. 44, 4678–4734.

Leliveld, J., Dentener, F.J., 2000. What controls tropospheric ozone? J. Geophys. Res. 105, 3531–3551.

Liu, S.C., Trainer, M., Fehsenfeld, F.C., Parrish, D.D., Williams, E.J., Fahey, D.W., Hübler, G., Murphy, P.C., 1987. Ozone production in the rural troposphere and the implications for regional and global ozone distributions. J. Geophys. Res. 92, 4191–4207.

Lin, X., Trainer, M., Liu, S.C., 1988. On the nonlinearity of the tropospheric ozone production. J. Geophys. Res. 93, 15,879–15,888.

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

Oltmans, S.J., Lefohn, A.S., Harris, J.M., Galbally, I., Scheel, H.E., Bodeker, G., et al., 2006. Long-term changes in tropospheric ozone. Atmos. Environ. 40, 3156–3173.

Parrish, D.D., Millet, D.B., Goldstein, A.H., 2009. Increasing ozone concentrations in marine boundary layer air inflow at the west coasts of North America and Europe. Atmos. Chem. Phys. 9, 1303–1323.

Sander, R., Baumgaertner, A., Gromov, S., Harder, H., Jöckel, P., Kerkweg, A., Kubistin, D., Regelin, E., Riede, H., Sandu, A., Taraborrelli, D., Tost, H., Xie, Z.-Q., 2011. The atmospheric chemistry box model CAABA/MECCA-3.0. Geosci. Model Dev. 4, 373–380. doi:10.5194/gmd-4-373-2011.

Stevenson, D.S., Dentener, F.J., Schultz, M.G., Ellingsen, K., van Noije, T.P.C., Wild, O., Zeng, G., et al., 2006. Multimodel ensemble simulations of present-day and near-future tropospheric ozone. J. Geophys. Res. 111, D08301. doi:10.1029/2005JD006338.

Solomon, S., Gian-Kasper Plattner, G.-K., Knutti, R., Friedlingstein, P., 2009. Irreversible climate change due to carbon dioxide emissions. Proc. Natl. Acad. Sci. U. S. A. 106, 1705–1709.

Uherek, E., Halenka, T., Borken-Kleefeld, J., Balkanski, Y., Berntsen, T., Borrego, C., Gauss, M., Hoor, P., Juda-Rezler, K., Lelieveld, J., Melas, D., Rypdal, K., Schmid, S., 2010. Transport impacts on atmosphere and climate: land transport. Atmos. Environ. 44, 4772–4816.

Wang, Z.S., Chien, C.-J., Tonnesen, G.S.J., 2009. Development of a tagged species source apportionment algorithm to characterize three-dimensional transport and transformation of precursors and secondary pollutants. J. Geophys. Res. 114, D21206. doi:10.1029/2008JD010846.

Wild, O., Prather, M.J., Akimoto, H., 2001. Indirect long-term global radiative cooling from $NO_x$ emissions. Geophys. Res. Lett. 28 (9), 1719–1722. doi:10.1029/2000GL012573.