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**Geoscientific
Model Development
Discussions**

Interactive comment on "On the attribution of contributions of atmospheric trace gases to emissions in atmospheric model applications" *by* V. Grewe et al.

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Reply to Anonymous Reviewer # 1

We like to thank the reviewer for the constructive comments, which improved the mansucript. Note that we have indicated changes in bold in the revised manuscript.

General points:

Applicability to real chemistry This point has been addressed at page 828 lines 6-8, but, we admit, not in a detailed way. We revised this part:

For applications in real chemistry schemes, the tagging method is in principle not dif-

ferent from the described one. To each species, n (number of regarded emission categories) tagged species are associated. For each of this tagged species production and loss terms have to be deduced. This decomposition of the production and loss terms into the contributions from individual emission categories is essential to the tagging methodology. This is a combinatorical problem, which can be solved in analogy to the above mentioned cases for 2 and 3-body reactions. A general approach is given in Appendix B. However, since the tagging of a whole chemical system is likely to be too computational demanding a mapping of the complex chemical system, including the production and loss terms, onto a simpler family concept might be helpful. Then only the families need to be tagged (Grewe, 2004).

Sensitivity method inappropriate We agree with the statement that the "Sensitivity method is inappropriate for source attribution" and have added a sentence in Section 6 "Implications and recommendations for attribution studies", last paragraph:

"To summarize, it has to be noted that the sensitivity method is in principle inappropriate for source attribution, but well suited to address impacts of e.g. future emission policies."

And in the conclusions:

Note, that the sensitivity method is, based on its concept, inappropriate for source attribution, but well suited to address impacts of e.g. future emission policies. However, since neither a full tagging of the modelled chemistry schemes, nor a tagging system with interrelationships between NO_x an VOC emissions has been implemented in models, we see the need for a further use of the sensitivity method, though inapproriate for source attribution. In order to assess some principle short-comings, we have introduced two error calculations (see below), which we recommend.

Specific comments Intro, 2nd Para We agree and included the following paragraph after the 2nd par. Obviously, for species, which are controlled by linear processes, like ^{222}Rn or SF_6, both approaches will lead to identical results (Fig. 1b). For non-linear

systems both methods might deviate only little (Fig. 1c) if the approximated tangent and the origin line differ only slightly. However, as soon as the system becomes non-linear differences between the approaches have the potential to increase largely (Fig. 1c).

Section 3, 1st para This point has been adressed in the introduction and conclusion. Here the focus of the paragraph is different, namely the uniqueness of the solution. We have added a line in the 1st paragraph of the introduction to stress the reviewer's point.

R12/13 Right! However, the assumption holds for the whole chemistry. The tagging formalism follows the assumptions of the resolved chemistry itself. Sub-grid processes may be resolved with plume approaches, for which the tagging then has to be extended. However, we think that this is beyond the scope of this paper.

Weaknesses Right! There are possibilities to account for the errors from transport schemes and at least to calculate the impact, e.g. as described in Grewe (2006). However, there are transport schemes, which are mass conserving by definition, e.g. Lagrangian methods, like ATTILA, which will become more and more popular, when the number of transported species increases, since the computational effort is almost inaffectes with these approaches. We propose to discuss this in an upcoming paper, which describes a taggig for a 3D chemistry scheme.

p.828, l.10 Agreed, but I think we have made this point now clear in the intro. and conclusion.

p.831, line 16 Changed - thanks.

p.835, l.3 ok

p.836, l.2 The conventions are checked. There are many reasons for this behaviour. The basic question is in how many cases the calculated derivative is closer to the line (0,0) (x,f(x)).

p.839, l.2 Wu and Fiore added.

p.839, l.12 Horowitz 1999 added.

Figure 1 splitted in 6 parts (see enclosed Figure):

1a General settings
1b Linear case and perfect agreement
1c Nonlinear case and good agreement
1d Nonlinear case and obvious problems
1e Details on calculation of contributions
1f Details on the two error definitions

Chemical Reactor Since both reviewer misunderstood the wording "chemical reactor", we changed it into "chemical system".

Interactive comment on Geosci. Model Dev. Discuss., 3, 819, 2010.

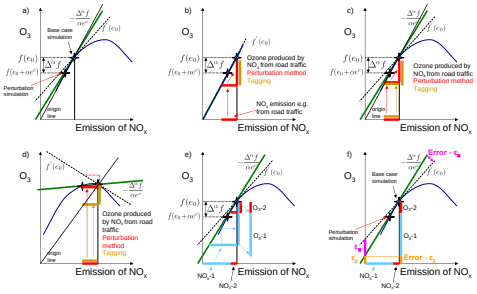

Fig. 1. Illustration of the sensitivity method (pair of simulation) to derive contributions from emission categories and intercomparison with the tagging method. The ozone concentration in arbitrary units is shown as a function of the emission of NO_x. Two simulations (base case and a simulation in which the emissions e_o is changed by a factor α) are indicated with stars. The derivative is added as a tangent for the base case (dashed line). The line through the base case simulation and the origin (origin line) is dotted. The green line shows the estimated derivative, based on the two simulations. a) General settings and calculation of the derivative. b) Assumption of linearity in ozone chemistry for illustration purpose. An arbitrary NO_x emission (horizontal red line) is considered. The vertical red and brown lines indicate the ozone contributions caused by this NO_x source (sensitivity method in red and tagging in brown) giving identical results. c) As b) but for the assumption of a non-linear ozone chemistry, however in a situation, which is close to the linear case. The green and dotted lines are used to calculate the contributions based on the sensitivity and tagging method, respectively. d) As c), but for a situation, which is far from the linear regime. e) Calculation of the ozone contributions; Two emission categories are considered (NO_x-1: light blue, NO_x-2: red) and the ozone contributions O_3-1 and O_3-2 indicated with vertical lines. f) Error analysis; The two errors ϵ_α (magenta) and ϵ_β (orange), which describe uncertainties associated with the determination of the tangent and the total estimate of all contributions (intersection of y-axis and tangent) (see Sect. 6).

Fig. 1.