**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.

Anonymous Referee #1

Received and published: 27 July 2010

This paper addresses the problem of source attribution for secondary atmospheric pollutants in numerical models, i.e., quantifying the contribution of different precursor emission sources to observed concentrations. This is a non-trivial problem, and there is as yet no consensus on how this should be done, and this paper makes a substantial contribution to helping resolve this issue. It describes the two key methods used (tagging vs. the sensitivity approach) and then presents a formal method for using the tagging approach, applying it to two stylized chemistry schemes with different underlying behaviors. This formalization is then used to quantify the errors that are associated with applying the sensitivity approach to assess source contributions. The study has been thoughtfully devised and the paper is well written, and as the topic is of consid-

erable interest to atmospheric modelers I believe that it is appropriate for publication in GMD with only minor revisions.

One issue that needs to be addressed in more detail is the applicability of the formal tagging method described here to real chemistry schemes involving more complex interactions. The two stylized schemes described are valid, but it is not clear that they adequately cover all situations possible in a real chemistry. A brief assessment of this would be very helpful. A second concern is that the paper implies that the sensitivity approach is appropriate for source attribution; while some studies in the past have used it this way, it would be helpful to point out that it is not an appropriate method for this purpose. A strength of the current study is that it allows quantification of the errors in this approach, but the authors should remind readers that the approach should be used for emissions sensitivities only, not for source attribution.

Specific Comments

Intro, 2nd Para: It would be useful to acknowledge simpler, linear cases (e.g., Radon, CFCs and other primary pollutants) where the two approaches give the same answer, before highlighting the differences for secondary pollutants, particularly those with non-linear chemistry such as ozone. The focus of this paper on secondary species such as ozone is implicit, but should be stated here explicitly.

Section 3, 1st para: A more important question here is why the source contribution matters. The main reason is that the climate impacts depend on the absolute contribution from a particular source. This should be clearly differentiated from the key motivation of the sensitivity approach which is to determine the effects of emission changes around some standard conditions representing the present, i.e., the local gradient term.

R12/R13: This makes the assumption that conditions are well-mixed. This is fine for stylized (or real) chemistry at a single point/box, but does not apply in the real atmosphere where there may be preferential reaction of Z_i or Z_j driven by heterogeneity in species distribution. What impact would this be expected to have?

There are weaknesses in the tagging methods as applied in 3-D models which are not acknowledged here. No model advection scheme is sufficiently conservative that the sum of constituent components equals the mass of total tracer, even for inert tracers. The absence of a practical implementation of the tagging approach in current models is acknowledged at the end of section 6, but other weaknesses in the approach should also be mentioned here.

p.828, l.10: Note that this is misuse of the sensitivity method. It should not be used for source attribution in this way (but sometimes is)

p.831, line 16: "X=10*Y" should read "X=Y/10" both here and in the legend to Fig 3.

p.835, l.3: perhaps add "even for the simpler chemistry considered here"

p.836, l.2: Fig 1 (and intuition) would suggest that the errors should be greater for positive perturbations than negative ones based on the curvature of the function shown. Some additional explanation is needed here. Please check the sign convention used for alpha is consistent here with p.828.

p.839, l.2: These seasonal variations in the errors are clearly illustrated in S. Wu et al., GRL, 2009 (Fig 2), and also in Fiore et al., JGR, 2009 (Fig 9).

p.839, l.12: I would expect some reference to Horowitz et al., JGR, 1999, as an example of an attempt at a reasonably full (yet not complete) tagging scheme applied in a large-scale model.

Figure 1 is very helpful but needs simplification as it provides too much information at once. It would be useful to separate the annotation (blue, red and orange bits) from the basic graph showing the function and gradient terms, so the figure could be presented in two panels to highlight these separate features. Showing the gradient line (f-prime) as a dashed line would also improve clarity.

Minor grammar and style issues, etc.

p.820, l.2: 'revised' should be rephrased as 'improved' or 'extended'?

p.821, l.6: remove 'more'

p.821, l.22: 'educts' -> 'reactants'

p.824, l.19: remove 'how'

p.825, l.12: 'reactors' -> 'reactants'

p.825, l.16: 'requested' -> 'required'

p.830, l.12: 'affin' -> 'affine' (it would be better to remove this and simplify the sentence.

p.830, l.17: 'provide a possibility' -> 'demonstrate'

p.831, l.12: 'increasing stronger' -> 'increases more strongly'

p.831, l.12: 'rich NOx' -> 'NOx rich'

p.835, l.16: add 'all except a' between 'for' and 'very'

p.835, l.5: 'second of' -> 'second on'

p.835, l.6: 'inequation (42) from an equation' perhaps clearer as 'the expression (42) from equality'

p.837, l.12: 'largely' -> 'greatly'

p.838, l.6: replace with 'how far the expressions (70) and (71) deviate from equality'

p.838, l.11: 'existent' -> 'included', 'reactors' -> 'reactions'

p.838, l.14: Sentence starting 'Note that...' needs revising as the meaning is not clear here.

p.839, l.5: Rephrase sentence: replace 'similar to' with 'like' and 'reactor' with 'scheme'?

p.839, l.12: 'Blow up' is too colloquial, please rephrase.

p.842, line before R14: '{1,...,M}' should be '{1,...,m}'

p.842, B2 and B3: check the formatting on these equations.

p.843, l.16: 'Prietly' -> 'Priestly'