**Geoscientific
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A generalized tagging method

V. Grewe

DLR-Institut für Physik der Atmosphäre, Oberpfaffenhofen, 82234 Wessling, Germany

Correspondence to: V. Grewe (volker.grewe@dlr.de)

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Abstract

The understanding of causes of changes in climate-chemistry simulations is an important, but often challenging task. In atmospheric chemistry, one approach is to tag species according to their origin (e.g. emission categories) and to inherit these tags
to other species during subsequent reactions. This concept was recently employed to calculate the contribution of atmospheric processes to temperature. Here a new concept for tagging any state variable is presented. This generalized tagging method results from a sensitivity analysis of the forcing terms of the right hand side of the governing differential equations. In a couple of examples, the consistency with previous
approaches is shown. Since the method is based on a ratio describing relative sensitivities, singularities occur where the method is not applicable. For some applications, like in atmospheric chemistry, these singularities can easily be removed. However, one theoretical example is given, where this method is not applicable at all.

1 Introduction

In order to answer questions like: "what is the contribution of road traffic emissions to climate change?" and "what is the contribution of anthropogenic CO_2 emissions to climate change?" climate (-chemistry) models were applied to provide an answer (Uherek et al., 2010; IPCC, 2007). Most applications rely on the assessment of changes, i.e. how sensitive reacts the atmosphere to a change in a regarded quantity, like road traffic emissions. Such a comparison of two experiments, one including all emissions, processes, etc. and one where the regarded process is altered (e.g. road traffic emissions suppressed), provides a valuable information on the sensitivity of, e.g. the atmosphere to road traffic emissions. However, it was shown that this, so-called perturbation approach does not provide a reliable estimate of the contribution of these emissions to ozone and climate change (Wang et al., 2009; Grewe et al., 2010, 2012; Emmons et al., 2012). For example, when ozone production is saturated, i.e. additional NO_x molecules only

lead to to a very low additional ozone production, this very low ozone production is applied in the perturbation method to estimate the total ozone produced.

On the contrary, the tagging methodology provides a useful framework to obtain information on the contribution of individual processes to specific quantities. It is widely used in atmospheric chemistry, but only recently well documented (Grewe, 2004, 2012; Wang et al., 2009; Gromov et al., 2010; Grewe et al., 2010; Butler et al., 2011; Emmons et al., 2012). In most applications, a subset of species is regarded and tagged without non-linear interaction between the individual species. For example road traffic emissions and the contributions to nitrogen oxide and ozone concentrations are tagged, but the interaction of, e.g. road traffic nitrogen oxides emissions and ship traffic non-methane hydrocarbon emissions is not regarded. Grewe et al. (2010) provided a methodology, which allows a complete tagging of a chemical scheme and takes these non-linear inter-dependencies into account.

Recently, this methodology was applied to a simple climate-box model to investigate the impact of atmospheric absorption on surface temperature, i.e. the greenhouse effect (Grewe, 2012). Hence it shows that in principle the tagging methodology is applicable to also other quantities than chemical species, only.

Here, these approaches are generalized to provide a framework with which any quantity can be tagged. This generalized tagging approach is introduced in Sect. 2. Section 3 provides four examples, which show how this formalism can be applied. These examples prove the consistency with previous approaches in Grewe et al. (2010); Grewe (2012), which were derived with a different, combinatorial, ansatz. The applicability of this methodology is limited by the existence of singularities, which are explored in more detail in Sect. 4.

2 A generalized tagging method

In a very generalized form, climate-chemistry models describe the temporal development of n state variables x_i, $i = 1,..,n$, which can be written in vector form: $\boldsymbol{x}^\mathrm{T} =$

$(x_1, \ldots, x_n) \in \mathbb{R}^n$. This temporal evolution is given by differential equations, describing the dependence on external forcings $\boldsymbol{P}(t)^{\mathsf{T}} = (P_1(t), \ldots, P_n(t)) \in \mathbb{R}^n$ and on the state variables themselves $\boldsymbol{F}(x)^{\mathsf{T}} = (F_1(x), \ldots, F_n(x)) \in \mathbb{R}^n$:

$$\frac{\partial}{\partial t} \boldsymbol{x} = \boldsymbol{P}(t) + \boldsymbol{F}(x). \tag{1}$$

5   In general, $\boldsymbol{F}(x)$ describes a sum of individual processes, like various reactions. Without loss of generality, this is restricted to one process only. The method described below can then be applied to each summand individually.

Now, we are interested in following the contributions of individual processes or quantities to the state variables. For example in atmospheric chemistry applications the contribution of emissions (here $\boldsymbol{P}(t)$) to the atmospheric composition (here x) is of interest. Another example is climate modelling: the contribution of greenhouse gases on temperature. Hence we define m categories, which totally partition the right hand side and, as a consequence also the left hand side of Eq. (1):

$$P_i(t) = \sum_{j=1}^{m} P_i^j(t) \tag{2}$$

$$F_i(x) = \sum_{j=1}^{m} F_i^j(x). \tag{3}$$

$$x_i = \sum_{j=1}^{m} x_i^j \tag{4}$$

Obviously, the challenging part of the decomposition is to derive the terms $F_i^j(x)$. In atmospheric chemistry a combinatorial approach was chosen to derive these terms (Grewe et al., 2010). That means that for a reaction of, e.g. species x_1 with species x_2, every possible combination of the categories j and k of either species was calculated.

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Hence all possible combinations of the contributions x_1^j and x_2^k of species x_1 and x_2 were calculated. Here, a different approach is chosen. In Sect. 3 it is shown that both approaches are equal and lead to the identical results.

The basic question is: "what impact has category j on the term $F_i(x)$?". The impact is defined as the sensitivity of the category j on the right hand side multiplied by state variable of category j. Or in other words, the impact Q_i^j of category j for the term F_i is:

$$Q_i^j = \boldsymbol{x}^j \frac{\partial F_i(x)}{\partial \boldsymbol{x}^j}, \tag{5}$$

where $\boldsymbol{x}^j = (x_1^j, \ldots, x_n^j)^\mathsf{T}$, the contribution of category j to x. The total impact T_i of all categories is then

$$T_i = \sum_{j=1}^{m} Q_i^j. \tag{6}$$

And the contribution $F_i^j(x)$ of the category j to $F_i(x)$ is the relative contribution of category j:

$$F_i^j(x) = \frac{Q_i^j}{T_i} F_i(x) \tag{7}$$

The differential equations for the tagged quantities x_i^j are then:

$$\frac{\partial}{\partial t} x_i^j = P_i^j(t) + \boldsymbol{F}_i^j(x) \tag{8}$$

$$= P_i^j(t) + \frac{Q_i^j}{T_i} F_i(x). \tag{9}$$

In vector notation and with $\frac{\partial x}{\partial x_i} = \frac{\partial \sum_{j=1}^{m} x_j}{\partial x_i} = \frac{\partial x_i}{\partial x_i} = 1$ follows:

$$\frac{\partial}{\partial t} x_i^j = P_i^j(t) + F_i(x) \frac{x^{j^\mathsf{T}} \nabla F_i(x)}{x^\mathsf{T} \nabla F_i(x)}. \tag{10}$$

Obviously, the method has a singularity and hence needs special treatments for situations where $x^\mathsf{T} \nabla F_i(x) = 0$. Before this singularity will be discussed in more detail, examples will be given to obtain a better understanding of the practical consequences of Eq. (10) and the consistency with previous tagging approaches in Grewe et al. (2010); Grewe (2012) will be shown.

3 Examples

3.1 Self-dependency

In many cases the regarded quantity x_i depends only on a temporal forcing and on itself, i.e. $F_i(x) = (0, \ldots, 0, f(x_i), 0, \ldots, 0)^\mathsf{T}$:

$$\frac{\partial}{\partial t} x_i^j = P_i^j(t) + f(x_i) \frac{x_i^j f'(x_i)}{x_i f'(x_i)} \tag{11}$$

$$= P_i^j(t) + f(x_i) \frac{x_i^j}{x_i} \tag{12}$$

This means that the right hand forcing term is linearly decomposed into the contributions according to the contributions of the state variable x_i^j.

Examples are tracers, like Radon, which are emitted at the Earth's surface (P_i^j) and decay radioactively, i.e. $f(x_i) = -\frac{x_i}{\tau}$, with τ the lifetime of the regarded species.

3.2 Bimolecular reactions and similar processes

Here a reaction of two species is regarded:

$$x_1 + x_2 \longrightarrow x_3, \tag{R1}$$

e.g. the reaction of NO and HO_2, which forms OH and NO_2 and latter photolyses and recombines to O_3. Therefore, we consider three species only, i.e. $n = 3$. The differential equation for the production of x_3 by this equation is

$$\frac{\partial}{\partial t} x_3 = F_3(x) = k x_1 x_2, \tag{13}$$

with k the reaction rate coefficient.

In this case $F_3(x) = k x_1 x_2$ with the notation of Eq. (10). With j categories the differential equation for the tagged species x_3^j becomes:

$$\frac{\partial}{\partial t} x_3^j = F_3(x) \frac{(x_1^j, x_2^j, x_3^j)^{\mathsf{T}} (k x_2, k x_1, 0)}{(x_1, x_2, x_3)^{\mathsf{T}} (k x_2, k x_1, 0)} \tag{14}$$

$$= F_3(x) \frac{k x_1^j x_2 + k x_2^j x_1}{2 k x_1 x_2} \tag{15}$$

$$= F_3(x) \frac{1}{2} \left(\frac{x_1^j}{x_1} + \frac{x_2^j}{x_2} \right) \tag{16}$$

Therefore the contribution of the category j to the production of x_3^j is determined by the mean contribution of the educts, i.e. $1/2(x_1^j/x_1 + x_2^j/x_2)$. Hence it is identical to results in Grewe et al. (2010).

3.3 Ternary and multi-body reactions

Let us now consider a chemical reaction, where $(m-1)$ educts lead to the species x_m:

$$x_1 + \ldots + x_{m-1} \longrightarrow x_m. \tag{R2}$$

The differential equation for the production is then:

$$\frac{\partial}{\partial t} x_m = F_m(x) = k_m \prod_{i=1}^{m-1} x_i \tag{17}$$

with k_m the reaction rate coefficient.

The derivative of the right hand side gives:

$$\frac{\partial}{\partial x_i} F_m(x) = k_m \prod_{k=1, k \neq i}^{m-1} x_k = \frac{F_m(x)}{x_i}. \tag{18}$$

And therefore, according to Eq. (10), the differential equation for the contribution results in

$$\frac{\partial}{\partial t} x_m^j = F_m(x) \frac{\sum\limits_{i=1}^{m-1} x_i^j \frac{\partial}{\partial x_i} F_m(x)}{\sum\limits_{i=1}^{m-1} x_i \frac{\partial}{\partial x_i} F_m(x)} \tag{19}$$

$$= F_m(x) \frac{\sum\limits_{i=1}^{m-1} \frac{x_i^j}{x_i} F_m(x)}{\sum\limits_{i=1}^{m-1} F_m(x)} \tag{20}$$

$$= F_m(x) \frac{1}{m-1} \sum_{i=1}^{m-1} \frac{x_i^j}{x_i} \tag{21}$$

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This means that the contribution of category j to the production of a species x_m by a $(m-1)$-body chemical reaction is the mean of the contributions of the individual species (educts) x_i to category j. This is again consistent with the previously derived equations for, e.g. a ternary reaction (Grewe et al., 2010).

3.4 Heating rates

Here an example is given for an alternative numerical approach to calculated the right hand side of the tagging equation Eq. (10). Consider tagging of temperatures (Grewe, 2012). The temperature equation includes diabatic terms and one of them is the diabatic heating rates from radiative processes. Instead of going through the math in the radiation calculation to derive the right hand side of the tagging equations, it is also possible to additionally calculate the heating rates with perturbations for each of the m categories.

Assume that the radiative transfer code provides vertical profiles in heating rates $H(z)$ (in $\mathrm{K\,s^{-1}}$) for an altitude coordinate z at any given location. For simplicity reasons, we assume only one state variable temperature T and two categories, which influence temperature, greenhouses gases (category 1) and solar influx and shortwave absorption (category 2).

The radiation code is then called additionally twice for every "perturbation"-altitude z_p with a perturbation in the temperature and with a perturbation in the greenhouse gas concentration (in ppbv). This provides a change in the heating rate profile per change in temperature and greenhouse gas concentration, respectively at all perturbation altitudes: $H_T(z, z_p)$ in $\mathrm{s^{-1}}$ and $H_G(z, z_p)$ in $\mathrm{K\,s^{-1}\,ppbv^{-1}}$.

The heating profiles $H^j(z)$ for the individual categories j depend on both the contributions of the two categories to the temperature $T^j(z)$ and the greenhouse gas concentration $G^j(z)$:

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$$H^j(z) = H(z) \frac{\sum\limits_{z_p} \left(H_T(z,z_p) T^j(z_p) + H_G(z,z_p) G^j(z_p) \right)}{\sum\limits_{z_p} \left(H_T(z,z_p) T(z_p) + H_G(z,z_p) G(z_p) \right)}. \tag{22}$$

Note that the magnitude in the perturbation has to be considered carefully in order to avoid impacts from numerical noise, if the perturbation was chosen to small and to avoid a too large deviation from the derivative if the perturbation was chosen too large.

4 Singularities in the tagging formula

The tagging method as described in Eq. (10) has an obvious disadvantage, since it may not be applicable in situation where $x^T \nabla F_i(x)$ becomes zero. Fortunately, this situation does not occur for many applications, e.g. for atmospheric chemistry or simple temperature tagging, as shown in the examples in Sects. 3.1–3.3. However, some singularities remain in these examples, namely if the state variables and concentration of species become zero. In these cases it can be argued that since no reaction occur, the contributions of the tagged species to the individual species remains unchanged.

In general, $x^T \nabla F_i(x)$ means that the sensitivities of the term F_i are fully balanced. Figure 1 gives an example for a two-dimensional situation, with two state variables x_1 and x_2. Here, the forcing term (thin isolines) is $F_1(x) = 0.25 - (x-a)^2$, with $a^T = (1/2, 1/2)$. Hence the derivative is $\nabla F_1(x) = -2(x-a)$. For every x with $x^T \nabla F_1(x) = 0$ (thick line) the sensitivity of the forcing term $F_1(x)$ with respect to x_1, which is $x_1 \frac{\partial}{\partial x_1} F_1(x)$, is balanced by the sensitivity with respect to the second variable x_2 and the sum equals zero. Or in other words x is perpendicular to $\nabla F_1(x)$ (arrows).

If we replace the forcing term by $F_1(x) = x_1/x_2$ in this example, we obtain an extreme singularity. For this forcing term the denominator in the tagging equation becomes zero:

$$x^\mathsf{T} \nabla F_1(x) = (x_1, x_2) \left(\frac{1}{x_2}, \frac{-x_1}{x_2^2} \right)^\mathsf{T} \tag{23}$$

$$= \frac{x_1}{x_2} - \frac{x_1}{x_2} = 0 \tag{24}$$

Hence, for individual cases, like atmospheric chemistry, singularities can consistently be removed, by setting the respective forcing term to zero, with the argument that if no reaction occurs also no changes in the tagged species can occur. However, as the last extreme case shows, theoretically, there are constellations, where an application of the tagging formula is not possible.

5 Conclusions

In this study, a tagging approach is presented, which allows to calculate contribution from individual processes or quantities on state variables for application in climate-chemistry models. For these contributions, also called tagged quantities or species, differential equations are presented, which result from a partitioning of the differential equations of the primary – untagged – state variables. This partitioning is based on the sensitivity of the right hand side forcing term with respect to the state variables.

This methodology is fully consistent with previous approaches presented in Grewe et al. (2010, 2012), which were derived with a different ansatz, a combinatorial approach. Four examples show how to apply this formalism to climate-chemistry applications. One example shows how the sensitivities can be derived numerically based on model internal sub-routines which were, in this case the radiation code, which provides heating rates.

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Despite the large possibilities, which this methodology offers, there are limitations to its applicability, since it may include singularities. For some applications, like for chemical tagging, the singularities arise from reactions, in which species are involved, which totally vanish. Since this implies that the reaction does not occur anymore, the right hand side of the tagging equation can be set to zero. This means that the involved tagged species remain unchanged, as the untagged primary species.

In a further theoretical example no consistent removal of these singularities could be found and in an extreme case the tagging formula is not applicable at all, since it consists of singularities, only.

Therefore, the examples show that this generalized tagging method is nicely applicable to a number of variables, like atmospheric concentrations or temperature. The limitations show that a careful consideration of the possible singularities is necessary, when applying the tagging formula.

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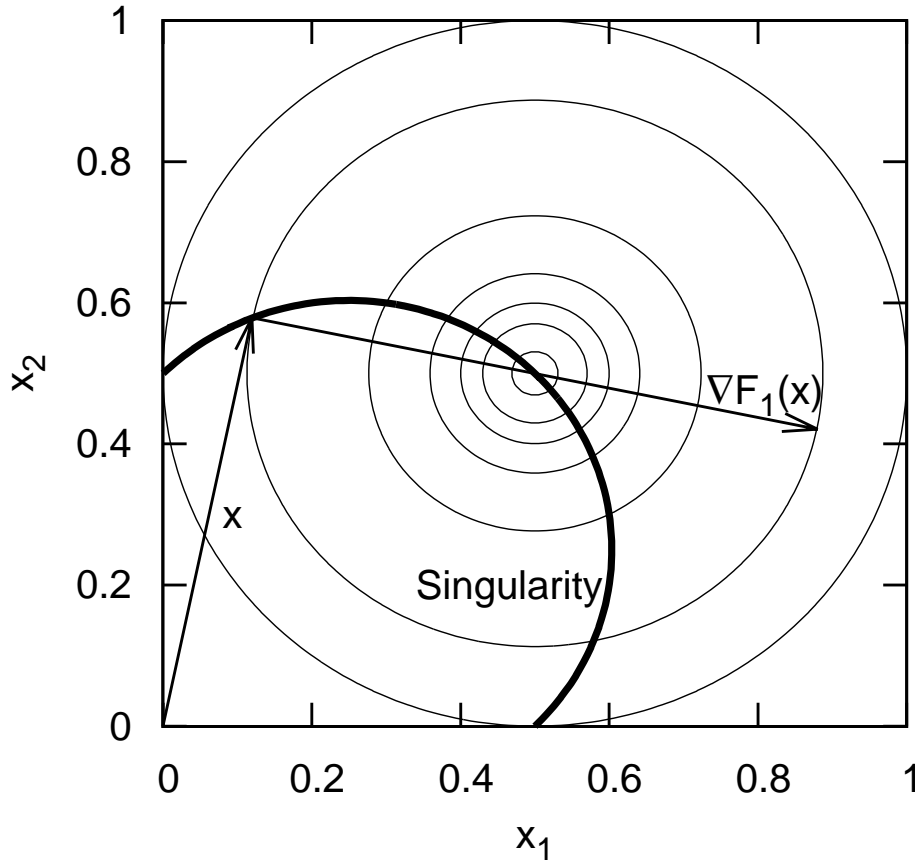
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Fig. 1. Example of a singularity in the tagging method. The forcing term $F_1(x)$ is shown in thin lines with isolines 0.,0.1, 0.2, 0.23, 0.24, 0.245, and 0.249 with a maximum in the middle. Thick lines indicate the location of the singularities $x \nabla F_1(x) = 0$. The arrows give an example.

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