Response to RC3

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For the convenience of the reviewer, we have included the comments from Reviewer 3 in black, normal font style and our responses indented in green, normal font style. The changes made to the manuscript or supplemental information corresponding to our response are provided in green, bold style font with the context of line numbers from the original manuscript and the original, remaining text in the normal font style.

This manuscript describes the theoretical base and implementation of the new CMAQ-hyd model for calculation of the first and second order sensitivities. This model is based on the hyperdual step method implemented into the widely used CTM model CMAQ (US-EPA). The manuscript describes also the evaluation and testing of the model including performance tests. The work brings significant new scientific results in the area of air quality modelling. The hyperdual step method has been already known but its utilisation in the CTM models is novel and very beneficial for the scientific community. The paper is well structured and clearly written. The findings are generally well described, I have a few specific comments described below. All corresponding materials (model source code and testing data) are available which allow to reproduce the experiments. The topic of the paper fits very well to GMD scope and I recommend the manuscript for publication in GMD after a minor revision (see specific comments below).

- We would like to thank the reviewer for their favorable and helpful comments on our manuscript. We have addressed the specific comments that the reviewer provided and consider the manuscript to be strengthened through this revision.

Specific comments:

I. Comments to the manuscript:

l. 149, Abbreviation SI is not defined. You probably meant Supplementary Material. Please, use the full name in this first occurrence of the abbreviation.

- Thank you for the correction. We have revised the manuscript to mention the first occurrence of Supplemental Information (SI).

  Lines 149 to 150:

  A demonstration of several basic operations is provided in the **Supplemental Information (SI)** while a more detailed discussion of the mathematical properties of hyperdual numbers is given by Fike and Alonso (2011).

l. 151-153, 162: The relation of a1, a2, a12 and Hh is not clearly formulated. Please, try to reformulate to make this step more comprehensible to the reader.

- Thank you for highlighting the need for more clarification on applying the hyperdual step method. Reviewer 1 expressed similar concerns. To address both comments, we revised the

description of the multiplicative hyperdual perturbation in the text and the SI. The changes to Lines 151 to 159 and to Line 15 in the SI are shown below.

Lines 151-159:

Akin to the Taylor series expansion about the real value of *x* in the finite difference method, the method of ascertaining sensitivities through a perturbation in **hyperdual space is based on a Taylor series expansion in an orthogonal dimension of the number**. **Specifically**, a hyperdual number with unity in a_0 and unity in one in $a_1$and a_2, **is multiplied** with the independent variable of interest before operating on it. After model execution, a Taylor series expansion is applied to extract sensitivities. **For instance,** the hyperdual-step method is applied to a scalar function, $f(x)$, by multiplying x by a hyperdual number, H_h, where $H_h = 1.0 + h_1\epsilon_1 + h_2\epsilon_2$. **This product** results in:

$$\begin{aligned} f(xH_h) = f(x) &+ (xh_1\epsilon_1 + xh_2\epsilon_2)f'(x) \\ &+ \frac{1}{2!}(xh_1\epsilon_1 + xh_2\epsilon_2)^2 f''(x) \\ &+ \frac{1}{3!}(xh_1\epsilon_1 + xh_2\epsilon_2)^3 f'''(x) + \cdots \end{aligned} \tag{5}$$

where "…" represents higher order terms in the series. Eliminating all terms that are zero due to the definition of hyperdual numbers (Eq. 2) **leads to**

$$f(xH_h) = f(x) + (xh_1\epsilon_1 + xh_2\epsilon_2)f'(x) + x^2 h_1 h_2 \epsilon_{12} f''(x) \tag{6}$$

where $f(xH_h)$ is a hyperdual number.

The properties of hyperdual numbers (Eqs. 2–4) lead to two significant results. First, all terms in the Taylor series expansion with derivatives higher than second-order become zero because all values include ϵ_1^2, ϵ_2^2, or ϵ_{12}^2. Second, the real component is unchanged. **A more detailed expansion of terms can be found in Eq. S7 in the SI or in the original development of hyperdual numbers, following the multiplication rule between a hyperdual and a real number** (Fike and Alonso, 2011).

Line 15, SI:

$$\begin{aligned} f(xH_h) &= f\big(x(1.0 + h_1\epsilon_1 + h_2\epsilon_2)\big) \\ &= f(x + xh_1\epsilon_1 + xh_2\epsilon_2) \\ &= f(x) + (xh_1\epsilon_1 + xh_2\epsilon_2)f'(x) + \frac{1}{2!}(xh_1\epsilon_1 + xh_2\epsilon_2)^2 f''(x) + \frac{1}{3!}(xh_1\epsilon_1 + xh_2\epsilon_2)^3 f'''(x) + \cdots \\ &= f(x) + (xh_1\epsilon_1 + xh_2\epsilon_2)f'(x) + \frac{1}{2}\big(x^2h_1^2\epsilon_1^2 + 2x^2h_1\epsilon_1h_2\epsilon_2 + x^2h_2^2\epsilon_2^2\big)f''(x) \\ &= f(x) + xh_1\epsilon_1 f'(x) + xh_2\epsilon_2 f'(x) + x^2h_1h_2\epsilon_{12}f''(x) \end{aligned}$$

The Taylor series expansion of the multiplicative hyperdual perturbation (Eq. 5) is shown in Eq. S7.

l. 214-232: This paragraph is not well formulated and the explanations are slightly disarranged. E.g. utilisation of the forward and reverse mode in CMAQ is not explained clearly. Similarly, the

adjustments done in the CMAQ itself, CMAQ-adjoint, and CMAQ-hyd are not clearly distinguished and all paragraph needs multiple readings to comprehend meaning of it. Please, try to reformulate it in a more straightforward way to allow easy understanding also to reader which is not fully familiar with the details of the CMAQ internals.

- Thank you for pointing out the need to rearrange explanations from Line 214 to 232. We have reformulated the paragraph to make it more logical.

  Lines 214-232:

  Several source code alternations were made to reduce the complexity of development and overcome the numerical instabilities related to hyperdual calculations in CMAQ's treatment of aerosol **specifically within the inorganic thermodynamic module ISORROPIA (Fountoukis and Nenes, 2007; Nenes et al., 1998). For the simplicity of development, we applied a Fortran 90-compliant version of ISORROPIA to replace the original Fortran 77 version of ISORROPIA in CMAQ.**

  ISORROPIA, as a key component of the aerosol module in CMAQ, is called either in the forward or the reverse mode. The forward mode of ISORROPIA takes the sum of gas and aerosol species concentrations, along with the relative humidity and temperature, to determine the partitioning of **Aitken- and accumulation-mode** species across the gas and aerosol phases **in CMAQ. In the original CMAQ model, ISORROPIA is run in the forward mode without limiting the temperature and pressure of the simulation. The determination process of species concentrations involves an iterative method which sometimes is numerically unstable during iterations for upper layer cells with low temperature and pressure for sensitivity computations with the HYD. To increase the numerical stability of CMAQ-hyd, we implemented temperature and pressure constraints so that the forward-mode ISORROPIA is only called when the cell temperature exceeds 260 K, and cell pressure exceeds 20,000 Pa. A similar set of temperature and pressure limits was applied to the call of ISORROPIA in the adjoint of CMAQ (Zhao et al., 2020). These changes do not affect the species concentrations computed by CMAQ while ensuring that the sensitivity computation process is stable.**

  To calculate the dynamic equilibrium of coarse mode aerosol species with the gas phase (Pilinis et al., 2000; P. Capaldo et al., 2000), CMAQ employs the reverse mode of ISORROPIA. **The input to reverse mode ISORROPIA includes concentrations of aerosol species, relative humidity, and temperature, and it results in partitioned concentrations in the** solid, liquid, and gas phases. The reverse-mode solution leads to unrealistic sensitivities calculated by HYD when the aerosol pH is close to neutral. One previous study found that the reverse ISORROPIA fails to capture the actual behaviour of inorganic aerosol when the pH is close to 7 (Hennigan et al., 2015). **To ensure stability of the sensitivity calculations, the changes to the hyperdual components in the coarse mode dynamic equilibrium are ignored** when the pH of coarse mode aerosol is close to neutral, **which ensures that the real components are identical to the original model.**

l. 242-243: The definition of Cpm2.5 and Enox should be moved forward somewhere to line 238 before their utilisation.

- Thank you for noticing the mentioning of Cpm2.5 and Enox before they are formally defined. We have rearranged the paragraph. To address the comment of Reviewer #1, we have also made some additional changes to the paragraph.

Lines 236-250:

Here, **for the sake of illustration**, we consider the semi-normalised sensitivities of **time-averaged** output concentrations of **ground-level** $PM_{2.5}$ concentrations, $\boldsymbol{C_{PM_{2.5},c,r,l=0,t}}$, to input NO_x ($NO+NO_2$) emissions, $\boldsymbol{E_{NO_x,c,r,l,t}}$, averaged over time**, *t*, for any given cell as indicated by the column, *c*, and row, *r*.** First-order semi-normalised sensitivities, $s_{NO_x}^{PM_{2.5}}$, and second-order semi-normalised sensitivities, $s_{NO_x}^{(2)PM_{2.5}}$ exemplify sensitivities relevant to environmental decision makers (Eqs. **15–16**).

Semi-normalised sensitivities reduce the complexity of interpretation by providing sensitivities in the units of the concentration per percent change of emissions. The semi-normalised sensitivities also scale down the impact from cells with low emission rates, which is consistent with the concentration reduction that is realistic **to expect**. Similarly, the time-averaged, semi-normalised cross-sensitivity of $PM_{2.5}$ to both NO_x and monoterpene is denoted as $s_{NO_x,TERP}^{(2)PM_{2.5}}$, with E_{TERP} representing the emission of monoterpenes (Eq. **17**).

$$s_{NO_x,TERP}^{(2)PM_{2.5}} = \frac{\partial^2 \overline{C_{PM_{2.5},c,r,l=0,tPM_{2.5}}}\big|_t}{\partial \overline{E_{NO_x,c,r,l,t}}|_t \overline{E_{TERP,c,r,l,t}}|_t} \overline{E_{NO_x,c,r,l,t}}|_t \overline{E_{TERP,c,r,l,t}}|_t) \quad \textbf{(17)}$$

l. 252: Superscripts inc, dec, and orig - explicit description of the meaning of these abbreviations might be beneficial even the following example gives a hint.

- Thank you for your suggestion. We have added an additional description of the meaning of these abbreviations as shown below.

  Lines 251-252:
  where the subscripts *c*, *r*, and *l* represent the column, row, and layer; the subscript *t* represents the time from the start of the model run; and the superscripts *inc*, *dec*, and *orig* represent the initial perturbation direction **(i.e., increased, decreased, and original emissions, respectively)**.

l.261-262: The last sentence partially repeats the statement of the sentence on lines 257-258. Moreover, section 2.1 discusses the hyperdual method and its errors while errors of the central FDM are discussed in the section 1 (l. 69-88).

- Thank you for pointing out the repetition of statements in these lines. We have removed this sentence from the manuscript.

l.269-270: The formulation "The second-order sensitivity evaluation is between a hybrid hyperdual-finite-difference method (HYD-FDM) and the hyperdual-step method." seems to be unclear. The approach is explained in the following text but the sentence can confuse the reader at the beginning. Please, reformulate.

- Thank you for pointing out the confusing of statements in these lines. We have reformulated the sentence as shown below.

  Lines 268-271:

Although the FDM can be applied to compute second-order sensitivities in CMAQ, previous studies have shown that the results are noisy and highly dependent on the perturbation sizes (Zhao et al., 2020; Zhang et al., 2012). **In order to evaluate the second-order sensitivities computed by the HYD method, we adopted a hybrid hyperdual-finite-difference method (HYD-FDM).** The **HYD-FDM** sensitivity calculation is given by:

l.286-287: You state here the evaluation has been done with 50% perturbation but in l.254-255 you assert the perturbation used has been 125% and 75%, i.e. 25%. I may overlook something and it may represent a different perturbation. Please, either correct these numbers (in case of the mistake) or add better explanation or description of these numbers (in case they are correct from some reason).

- Thank you for your suggestion. We have applied the central difference method with perturbations in both the increasing (125%) and decreasing directions (75%). We have added the following line to better explain the perturbation for finite difference.

  Line 285-289:
  We evaluated the implementation of CMAQ-hyd by comparing the first-order sensitivities of various species in CMAQ calculated by HYD with a hyperdual-step perturbation described in Section 2.3 (HYD sensitivities) and FDM with a domain-wide emission perturbation (FD sensitivities). **The FD sensitivities were computed with the difference between a 25% increase and a 25% decrease in domain-wide emissions.** Overall, different HYD and FD sensitivities agree well, as evidenced by the close alignment of the points on the blue identity line, which represents perfect agreement, in most panels of Figure 2.

l. 293-316: The Fig. 3 seems to be poorly arranged as the overlapping points do not allow the comparison of individual results. My suggestion is to rearrange the Fig. 3 in a way which will allow to study better the behaviour of the FDM subtraction and truncation errors with decreasing of the perturbations and to assess the "convergence" of FDM to hyperdual results. Four separate graphs might work better than the current unified graph. You can have a better idea how to deal with it.

- Thank you for your suggestion about rearranging Figure 3 to make the description clearer. We have altered Figure 3 so that it depicts the four separate approaches (125%, 75% FDM; 110%, 90% FDM; 105%, 95% FDM; HYD) in the modelling domain on a map. We have removed the forward and backward finite differences (125%, 100%; 100%, 75%) and added an additional central-difference-based calculation (105%, 95%).
- From the maps in the new Figure 3, we can observe that the FDM does not “converge” to the hyperdual results when we decrease the perturbation sizes. Due to numerical noise inherent to CMAQ, the sensitivities calculated with runs with smaller perturbation sizes deviate farther from the hyperdual results. This result is also described in a previous work (Zhang et al., 2012).
- We have also changed Figure 4 accordingly to reflect the change in computational scenarios used.
- We have removed the original Figure S1 since it is similar to the new Figure 3 in the main manuscript.

Lines 317 to 349:

The FD sensitivities with the base case perturbation (125 %, 75 %) and **two** other perturbation size pairs (110 %, 90 %; **105 %, 95 %)** are shown in Fig. 3. The FDM sensitivities calculated with different perturbation sizes **are plotted on Fig. 3a, Fig. 3b, and Fig 3c., respectively. The FDM sensitivities exhibit similar behaviour to the HYD sensitivities over the continents. However, the** inconsistency among the sensitivities calculated by FDM with different perturbation sizes **over the ocean** (Fig. 3) suggests that the FD sensitivities heavily depend on the perturbation sizes. This result **demonstrates** the **relatively** low credibility of FD sensitivities, particularly for highly nonlinear relationships where the truncation errors could be large. **Notably, reducing perturbation sizes in the FDM did not lead to convergence with hyperdual sensitivities. This divergence may be attributed to the propagation of numerical noise from the model run to the calculated sensitivities as perturbation sizes decrease. This finding is consistent with the results in Zhang et al. (2012).** Our findings demonstrate the importance of using other methods, including the HYD, which are not prone to truncation or cancellation errors for **probing** nonlinear relationships in CTMs.

We also compared the spatial distribution of HYD sensitivities (Fig. 4a) against the average (Fig. 4b) and the range (Fig. 4c) of the FD sensitivities with **three** different perturbation sizes. Differences are evident between the HYD and the average FD sensitivities in central North Carolina and Tennessee as well as off the coasts of Georgia and South Carolina. The HYD predicts slightly negative sensitivities in North Carolina and Tennessee while the FDM predicts slightly positive values. The average FDM sensitivities off the coast of Georgia and South Carolina were noisy, with alternating positive and negative sensitivities, while the HYD sensitivities were much less noisy.

L. 393-394: According figures 6g and 6h, the second order sensitivities of PM2.5 to TERP (Fig. 6g) are mostly negative, while to APIN (Fig. 6h) are mostly positive.

- Thank you for pointing out the mistake in the manuscript. We have corrected the mistake in lines 393-394.

Lines 393-394:

On the other hand, the $s_{TERP}^{(2)\mathrm{PM}_{2.5}}$ (Fig. 6g) is mostly **negative**, while $s_{APIN}^{(2)\mathrm{PM}_{2,5}}$ (Fig. 6h) is mostly **positive**.

l. 445-446: The implementation of CVM is not higher. You probably meant the computational cost of this CVM based model.

- Thank you for pointing out the mistake in the manuscript. We have corrected the mistake in lines 445-446.

Lines 445-446:

For instance, the implementation of CVM in GEOS-CHEM **results in** a 4.5-**fold increase in computational overhead when compared to** the **standard** model (Constantin and Barrett, 2014).

l. 448-449: The sentence does not make sense. Probably words were left out after “are shown for the”.

- Thank you for pointing out the unclear sentence in the manuscript. We have rewritten the sentence to make it clearer.

  Lines 448-449:
  The total wall time **with different numbers of computational nodes used** for identical runs of the original CMAQ model and CMAQ-hyd **is displayed** (Fig. 8).

l. 450: You hide from the reader the number of processor cores/MPI processes. This fact is much more important than number of nodes. The 7rganization of the MPI processes to individual nodes can only influence the Infiniband (or another transport layer) overhead which is usually small in modern HPC system for such a type of tasks. Also, the extent of your configuration is important mainly for assessment of the parallelization efficiency with growing number of MPI processes involved as well as for memory demands of the model. Please, give the reader full information about your testing configuration. You can add this information into the Section 4. Of Supplements and give a reference to it here.

- Thank you so much for your request of the detailed configuration of computing nodes. We have provided the additional information in the SI and added a sentence in the main manuscript referring the readers to the SI.

  Line 450:
  The CMAQ-hyd and regular CMAQ runs were performed with 1, 2, 4, and 8 nodes on the supercomputing cluster. **The configuration of computing resources is detailed in Section 4 of the SI**.

  Lines 42-45, SI: **The runs using 1, 2, 4, and 8 nodes runs use 36 processors, 72 processors, 144 processors, and 288 processors, respectively. CMAQ delegates the computing job of the grid by partitioning in the horizontal plane. Each processor is tasked with executing the scientific processes for a set of columns before communicating the select information on the boundaries. One example of the subdivision of the horizontal plane by 1 node (36 processors) is shown in Figure S3. Figure S3a demonstrates the 100 by 80 horizontal grid of our modelling domain, and Figure S3b demonstrates the subdivision of computing tasks by 36 processors. Each processor is responsible for computations in rectangular squares drawn in dashed lines. The columns of CMAQ are decomposed into 36 MPI processes in the shape of 6 columns and 6 rows of MPI processes. The specific shape of MPI processes is: 6 columns by 6 rows for 1 node (36 processors), 12 columns by 6 rows for 2 nodes (72 processors), 12 columns by 12 rows for 4 nodes (144 processors), and 24 columns by 12 rows for 8 nodes (288 processors). We applied the same computational configuration for the regular CMAQ and the CMAQ-hyd runs during code profiling processes.**

  When the number of processors used increases, the number of grid cells along the boundaries requiring information exchange increases. Consequently, there is an increase in relative time consumed by the I/O processes when the number of processors used increases.

l.483: I have slight doubts you can use the word “validated” here, I would suggest “evaluated” or a similar word. My reasoning is methodological. The purpose of tests done in section 3.1 is to evaluate the correctness and accuracy of the hyperdual based implementation. You compare results of the new more precise method with an established less precise method (according the theory) and you get some differences. You attribute these discrepancies between FDM and hyperdual results only to the nonlinearity of the model but how can you be sure they are not caused also by another reason, e.g. some problem in the hyperdual implementation? Yes, I am also convinced, that it is the result of FDM errors due to nonlinearity of the model but it is not a formal proof. You give a good supporting arguments in the following parts of the section 3.1 and they very well support the trust in the correctness of the CMAQ-hyd model implementation. But I still would be careful to call it formally validation. The thorough evaluation of the behaviour of CMAQ-hyd and its comparison with FDM done in 3.1 shows that the differences are of expected properties what allows to trust this new model.

- Thank you so much for this valuable input. We validated the hyperdual calculations defined in the HDMod operator overloading library. Motivated by a similar request for clarity in this comment and another of Reviewer #1, we have added a sentence to Lines 215-216 to explain the validation against analytical derivatives with a testing framework developed by Pellegrini and Russell (2016).
- Indeed, the comparison between the hyperdual-step method and the finite difference method does not serve as a formal proof. The word ‘evaluated’ is a good fit for describing the relationship between sensitivities calculated by CMAQ with finite difference and CMAQ-hyd.

  Lines 215-216:
  Before being applied to CMAQ, the operator overloading library was separately **validated by comparing against analytical derivatives using a testing framework developed by Pellegrini and Russell (2016).**

  Line 483-486:
  We developed and validated the hyperdual-step module “HDMod”, which **is limited to analytically verifiable** mathematical operations **of hyperdual numbers**. This module can also be applied to other numerical models where first- and second-order sensitivities are of interest. We further **evaluated** the development of CMAQ-hyd against the FDM and FDM-HYD hybrid method to ensure the correctness of the implementation. During the **evaluation** process, …

l. 507: "..free from numerical noise.." - Only specific types of the numerical errors (truncation and subtractive cancellation errors) are eliminated by this method. Even they are the most important ones, I would suggest a more careful formulation here.

- Thank you for suggesting the specific types of the numerical errors that the hyperdual-step method can avoid. We have updated the manuscript accordingly.

  Lines 486-487: CMAQ-hyd **computes** sensitivities free from **truncation and subtractive cancellation errors**, **unlike** those calculated by the FDM.

Lines 506-507: In conclusion, we have developed and evaluated CMAQ-hyd, a novel, augmented model to compute first-order, second-order, and cross-sensitivities free from **subtractive cancellation and truncation errors** in CMAQ.

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