

Interactive comment on “Source apportionment and sensitivity analysis: two methodologies with two different purposes” by Alain Clappier et al.

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The manuscript discusses relationships between source sensitivity and source contribution for air quality model simulations involving non-linear chemistry. Hypothetical examples are provided that may help readers better understand the concepts involved. Although this work does not present new concepts and results, it does provide in-depth discussion of issues that have sometimes created confusion when air quality model results are used to develop public health policy. I support publication. I request the following clarifications. Lines 25-29: However, this study demonstrates that when the relationship between emissions and concentrations is non-linear, sensitivity approaches are not suitable to retrieve source contributions and source apportionment methods are not appropriate to evaluate the impact of abatement strategies. The phrases “not

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suitable” and “not appropriate” suggest that there should be a brightline, yes or no, test for when sensitivity and source apportionment methods can be used. Instead, I suggest using more subtle wording to explain that these methods will encounter limitations when applied to non-linear cases. When non-linearity is mild these limitations may be acceptable in the context of the other uncertainties inherent to complex models. In other words, source apportionment may provide useful information about model sensitivity, and vice-versa, for mildly non-linear cases. In the main manuscript, it would be useful to cite Koo et al. (2009) who present a detailed comparison of source apportionment and sensitivity in a 3-D PM model that shows which sensitivities are similar to apportionment, and which are not. Koo, B., Wilson, G.M., Morris, R.E., Dunker, A.M. and Yarwood, G., 2009. Comparison of source apportionment and sensitivity analysis in a particulate matter air quality model. *Environmental Science & Technology*, 43(17), pp.6669-6675.

R: We agree with the Reviewer and adapted the text in several instances to smooth the wording. The Koo et al. reference has been inserted in the text.

Lines 297 - 298: The decoupled direct method (DDM) is designed to estimate sensitivities to emission changes (Dunker et al., 1984; Dunker et al., 299 2002). The DDM explicitly calculates sensitivity, as opposed to making an estimate. If a sensitivity calculated by a specific model turns out to be imprecise, that is a limitation of how the DDM is implemented.

R: This has been changed in the text according to reviewer comment

Lines 303 - 319: This discussion might be understood to suggest that Taylor series expansions are the only way to use sensitivity coefficients computed using the DDM. Dunker (2015) showed how to use first-order sensitivity to determine source contributions to the difference between two model cases, e.g., to apportion the difference between the current atmosphere and natural conditions to specific human activities. Simon et al. used first order sensitivity to construct emission response surfaces. It

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would be useful to mention other ways in which sensitivity information can be applied. It would be useful to note that computing first-order sensitivity at several emission levels is a powerful alternative to high-order sensitivity. Dunker, A.M., 2015. Path-integral method for the source apportionment of photochemical pollutants. *Geoscientific Model Development*, 8(6), pp.1763-1773. Simon, H., Baker, K.R., Akhtar, F., Napelenok, S.L., Possiel, N., Wells, B. and Timin, B., 2013. A direct sensitivity approach to predict hourly ozone resulting from compliance with the National Ambient Air Quality Standard. *Environmental Science & Technology*, 47(5), pp.2304-2313.

R: We agree. Sensitivity can be obtained with DDM as first order derivatives but also from a brute force approach. We tried to clarify this in the text. We also inserted the useful references suggested by the Reviewer.

Lines 348 to 356: $\text{PPM} \rightarrow \text{PM}[\text{PPM}]$ $\text{NO}_2 + \text{NH}_3 \rightarrow \text{PM}[\text{NH}_4\text{NO}_3]$ $\text{SO}_2 + 2 \text{NH}_3 \rightarrow \text{PM}[(\text{NH}_4)_2\text{SO}_4]$ The fact that reactions in lines 349 and 350 are non-stoichiometric may cause confusion. Changing NO_2 to NO_x and SO_2 to SO_x may make it more apparent that these reactions are intended to be conceptual. The subsequent statements about the stoichiometry of these reactions in lines 354 to 356 are chemically inaccurate and should be re-worded. The reaction in line 348 seems not needed because no atmospheric reactions are involved for primary PM, it is simply emitted.

R: A set of chemical stoichiometric equations has been added to explain from the origin of our simplified equation system. We hope this now clarifies the issue.

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