

Geosci. Model Dev. Discuss., referee comment RC1
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Comment on gmd-2021-259

Anonymous Referee #1

Referee comment on "A quantitative decoupling analysis (QDA v1.0) method for the assessment of meteorological, emission and chemical contributions to fine particulate pollution" by Junhua Wang et al., Geosci. Model Dev. Discuss., <https://doi.org/10.5194/gmd-2021-259-RC1>, 2021

General comments

The authors describe a method for determining the contributions of different processes to PM_{2.5} formation and the couplings between the processes. The authors call this method Quantitative Decoupling Analysis (QDA) and apply the method to a haze episode in the Beijing-Tianjin-Hebei region from 17-28 February 2014. The manuscript is generally well-written.

However, there are three significant problems with the work:

* The QDA method is not new. This is the Factor Separation method introduced in 1993 and applied in later work. See references below.

* The emissions are constant throughout the simulation period, which is not realistic. As a consequence, the contribution of emissions to the PM_{2.5} concentration change is constant throughout the episode, and all the time-variation in the factors and couplings is driven by the meteorology.

* The authors consider the influence of three factors on PM_{2.5}, total emissions, chemistry, and meteorology, and indicate that their work provides valuable information to decision makers (lines 64-74). PM_{2.5} pollution episodes are driven by anthropogenic emissions and meteorology. Chemistry is a secondary factor that responds to emissions and meteorology but can be controlled by decision makers only by regulating the anthropogenic emissions. It would be much more relevant to decision makers if the authors had chosen biogenic emissions, anthropogenic emissions, and meteorology as the three factors. Then the full effect of anthropogenic emissions on PM_{2.5} during the episode would be apparent, rather than burying some of the effect in the chemistry factor. Also, the results would help determine if emergency anthropogenic emission controls during episodes would reduce PM_{2.5}, which is a goal of the authors' work (lines 71-72) but not a result of their work.

Specific comments

p. 3, lines 64-72. Decision makers can control anthropogenic emissions and possibly have

a minor impact on some biogenic emissions (e.g., types of trees planted in urban areas). Understanding the impact of meteorology on atmospheric concentrations is also important and useful to decision makers. But separating out the impact of chemical reactions does not help regulators reduce atmospheric concentrations. The chemistry factor is controlled by emissions and meteorology, so some (not all) of the chemistry factor represents the impact of emissions. The decision makers need to understand the full impact of the emissions, but that cannot be obtained from the factors that the authors chose.

p. 4, lines 89-91. Eq. 2 is incorrect. There should be a factor of 2 in front of the cross terms $\Delta x_1 \Delta x_2$, $\Delta x_2 \Delta x_3$, and $\Delta x_1 \Delta x_3$ in the group of second-order terms and other non-unity factors derived from the binomial coefficients in front of the cross terms in the group of third-order terms. The authors may not have used Eq. 2 and used only Eq. 3. However, if the authors actually used Eq. 2 in their calculations and analyses, they should verify that they used the correct equation, and, if not, the calculations and analyses must be re-done. In any case, Eq. 2 should be corrected.

p. 4, line 104. The interaction between emissions and meteorology is bi-directional. Higher temperatures increase evaporative emissions from gasoline vehicles, higher temperatures and greater sunlight increase isoprene emissions from plants, etc.

p.4, lines 106-112 and Table 1. There should be a more complete description of the simulations and what is different between simulation M1 and the other simulations. In particular for simulation M4, are the meteorological processes and emissions absent for the entire simulation? If so, what PM2.5 could there be in a grid cell other than the initial PM2.5 concentration, which is stationary in space because no meteorological processes are included? Is the PM2.5 concentration at the start of a specific time step taken from simulation M1, simulation M4 is run over that time step without including meteorology processes or emissions anywhere in the modeling domain, then a new PM2.5 concentration is obtained from simulation M1 for the next time step? What the authors did is very unclear.

p. 4, lines 111-114. Did the authors run the base simulation 6 times, each time with one of the "accompanying" simulations? That would effectively be 12 simulations. A simpler approach would seem to be running the base simulation once, recording the timesteps used, and then running each of the "accompanying" simulations once with the same timesteps used for the base case. That would reduce the number of simulations needed to 7.

p. 4, lines 119-120. There should be a detailed explanation of how IPR is applied to the results for each factor. It is unclear how this was done. Simulation M4 (C factor) does not contain emissions, so the chemistry will be different from that when emissions are present. Are the IPR results then meaningful for C?

pp. 3-5. The QDA method is not new. This is the Factor Separation method introduced by U. Stein and P. Alpert, Factor separation in numerical simulations, *J. Atmos. Sci.* 50, 2107-2115 (1993). Subsequently, Tao et al. applied the method to separate the contributions of area, mobile, and point source emissions to ozone and their interactions (Tao et al., Area, mobile, and point source contributions to ground level ozone: a summer simulation across the continental USA, *Atmos. Environ.* 39, 1869-1877 (2005)). The authors should not refer to QDA as a new method and should credit Stein and Alpert and Tao et al. by including their references in the manuscript.

p. 7, line 187. What is MBE? This is not defined in Table S1. MB is $-13.7 \mu\text{g}/\text{m}^3$ and ME is $42.1 \mu\text{g}/\text{m}^3$ (Table S2) so it cannot be either of those two statistics.

p. 7, line 191. Again, what is MBE and where are these values (7.1 and $5.3 \mu\text{g}/\text{m}^3$) in

Table S2? If the values are discussed in the manuscript, they should be in Table S2.

p. 7, line 194. There is a more recent paper (L. Huang et al., Atmos. Chem. Phys. 21, 2725-2743 (2021)) that gives goals and criteria specifically for PM 2.5 simulations in China.

p. 7, lines 210-212. For their analyses, the authors fixed the emissions to be constant in time. It is unclear why this is necessary for the method, and it is a serious limitation of their work. Neither the anthropogenic nor the biogenic emissions are constant in time; there are large variations over the diurnal cycle. As a consequence of the authors' assumption of constant emissions, their calculated emission contribution is constant over all 12 days of the episode (Figures 7 and 8, Tables 3 and 4). This is not an interesting or very valuable result, especially for the decision makers/regulators. We cannot control the meteorology, only the anthropogenic emissions, so the important question is to what extent instituting greater emission controls during stagnation events will improve air quality. The authors' results do not provide any insight on that question. Further, the assumption of constant emissions also influences the chemical contribution because time-varying emissions would very likely give much greater variation in the chemistry contribution.

p. 8, lines 234-239. These conclusions are well-known from many previous studies.

p. 9, line 276. It is unclear what the range of -0.86 to 1.86 represents. It is much wider than what the results in Table 3 suggest.

pp. 10-11, Section 3.4. Many of the conclusions here are well-known from prior work, and the Factor Separation method (QDA) adds little new information to the prior work. At most, this section shows consistency between the Factor Separation method and the results of previous studies, but there is no detailed evaluation of the Factor Separation method.

p. 10, lines 281-282. Yes, the results in the paper do not give much information about the importance of emissions and therefore are not of much use to decision makers.

pp. 11-12. Again, QDA is not a new method and most of the conclusions here are not new.

Technical corrections

p. 2, lines 54-55. "However, due to the nonrepeatability of individual pollution cases," Not clear what is meant here. If one has an estimate of the meteorological fields from a weather model and an estimate of the emissions, the air quality model can estimate the atmospheric concentration of PM for days in different years ("individual cases"). Not clear why sensitivity experiments are necessary to "fully reproduce the individual cases."

p. 2, line 57. Define the PLMA acronym.

p. 5, line 133. Should be "nitrate" not "nitrite"?

p. 5, line 149. Was it MOZART v 2.4 or v 2.5?

p. 6, line 159. "The" should be "the"

Figure 2 (b). The legend should be larger.

p. 6, lines 179-180. It would be clearer to use the same nomenclature for these statistics as in Table S1. Table S1 has the conventional names.

Figure 6 caption. There is no solid line in the figure, only 3 dashed lines. Do the points represent 24 hour averages? For which days?

Tables 3 and 4 should be in the Supporting Information because they repeat the information in Figure 8.

p. 9, line 259. Not clear why one limit is -3%. This seems to be a comparison of the magnitudes of the two quantities, in which case the limit would be +3%.

p. 10, line 293. Should it be "from stages 1 to 2" instead of "from stages 2 to 1"?

p. 11, line 311. Define the acronym PLAM.

p. 14, lines 436-437. The title of the paper should not be in all capitals.