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Interactive comment on "Development of 3D Variational Assimilation System Based on GRAPES-CUACE Adjoint Model (GRAPES-CUACE-3D-Var V1.0) and Its Application in Emission Inversion" *by* Chao Wang et al.

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Interactive comment on "Development of 3D Variational Assimilation System Based on GRAPES-CUACE Adjoint Model (GRAPES-CUACE-3D-Var V1.0) and Its Application in Emission Inversion" by Chao Wang et al.

Anonymous Referee #2 Received and published: 22 May 2020

This manuscript presents a new chemical data assimilation system. While the work seems well motivated and potentially valuable eventually, overall the effort comes

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across as the first step in what needs to be a more thorough study prior to publication. From the onset, the manuscript struggles to explain why they are using an adjoint model for 3D-Var (typically this would be used for 4D-Var), which is then confused as they proceed to describe what is essentially a 4D-Var system but then provide code for something that seems like 3D-Var. . .it becomes very unclear what is the scope and objective. Further, the authors neglect to treat prior information in a Bayesian manner, leading to an ad hoc approach that is not justified. Other issues such as a lack of an appropriate dataset for testing, lack of cross validation, no exploration of the role of prior error covariances, and also the unavailability of the underlying model code amount to this work seeming rather premature. I look forward to a complete manuscript being submitted at a later date, here or elsewhere. Reply: We thank the reviewer for his/her careful reading of the manuscript and his/her valuable comments. We have deeply considered the points brought up by the reviewer, and have learned a lot from the comments. We did our best to upgrade the assimilation system, redesign the emission inversion experiment, obtain more observations, and improve the readability of the text. The introduction and methods have been rewritten as suggested. Unfortunately, we spent a long time in upgrading and debugging the assimilation system, so that we did not obtain more satisfactory results of emission inversion within the specified time. So in the revised manuscript, we are unable to present the results and conclusion, for which we are very sorry. The responses to the specific questions are shown below. Thanks again for the reviewer's valuable suggestions.

Main comments: The introduction briefly mentions many previous 4D-Var studies that have used adjoint models for estimating emissions of aerosols and trace gases. However, it is not discussed anywhere why the authors are using their adjoint model for 3D, what is the background of other studies using 3D-Var, and how this compares to 4D-Var. Nor is very much background provided about the GRAPES-CUACE model âAËŸT it isn't even ËĞ mentioned until the final paragraph. No background on model performance is discussed that motivates the need for the GRAPES-CUACE 3D-Var. In other words, the introduction is not written very specifically for this manuscript. Reply:

According to the reviewer's valuable suggestions, we have added the description of GRAPES-CUACE model and the motivation for the development of GRAPES-CUACE-4D-Var. We have rewritten this part as follow: "1 Introduction Three-dimensional (3-D) atmospheric chemical transport models (CTMs) are important tools for air quality research, which are used not only for predicting spatial and temporal distributions of air pollutants, but also for providing sensitivities of air pollutant concentrations with respect to various parameters (Hakami, et al., 2007). Among several methods of sensitivity analysis, the adjoint method is known to be an efficient means of calculating the sensitivities of a cost function with respect to a large number of input parameters (Sandu et al., 2005; Hakami et al., 2007; Henze et al., 2007; Zhai et al., 2018). The sensitivity information provided by the adjoint approach can be applied to a variety of optimization problems, such as formulating optimized pollution-control strategies, inverse modelling and variational data assimilation (Liu, 2005; Hakami, et al., 2007). Four-dimensional variational (4D-Var) data assimilation, which is an important application of adjoint models, provides insight into various model inputs, such as initial conditions and emissions (Liu et al., 2005; Yumimoto and Uno, 2006). In the past decades, many scholars have successively developed adjoint models of various 3-D CTMs and the 4D-Var data assimilation systems to optimize model parameters. Elbern et al. (1999, 2000, 2001, 2007) constructed the adjoint of the EURAD CTM and performed inverse modelling of emissions and chemical data assimilation. Sandu et al. (2005) built the adjoint of the comprehensive chemical transport model STEM-III and conducted the data assimilation in a twin experiments framework as well as the assimilation of a real data set, with the control variables being O3 or NO2. Hakami et al. (2005) adapted the adjoint model of STEM-2k1 model for assimilating black carbon (BC) concentrations and recovery of its emissions. Liu (2005) and Huang et al. (2018) developed the adjoint of CAMx model and further expanded it into an air quality forecasting and pollution-control decision support system. Müller and Stavrakou (2005) constructed an inverse modelling framework based on the adjoint of the global model IMAGES and used it to optimize the global annual CO and NOx emissions for the year 1997. More recently, the CMAQ commu-

nity (Hakami et al., 2007) built the adjoint of CMAQ model and its 4D-Var assimilation scheme, which were used to optimize NOx emissions (Kurokawa et al., 2009; Resler et al., 2010) and ozone initial state (Park et al., 2016). The adjoint of GEOS-Chem model and its 4D-Var assimilation system firstly developed by Henze et al. (2007, 2009) have been applied in a number of studies to improve aerosol (Wang et al., 2012; Mao et al., 2015; Jeong and Park, 2018), CO (Jiang et al., 2015) and NMVOC (Cao et al., 2018) emissions estimates. Zhang et al. (2016) applied the 4D-Var assimilation system using the adjoint model of GEOS-Chem with the fine 1/4°× 5/16° horizontal resolution to optimize daily aerosol primary and precursor emissions over North China. These researches have laid good foundations for developing adjoint models of CTMs and optimizing model parameters. However, only a few of these adjoint models and their 4D-Var assimilation systems have been widely applied to regional air pollution in China. The development and applications of adjoint models of 3-D CTMs and their 4D-Var data assimilation systems are still limited in China. Further research and more attention are required. Nowadays, several mega urban agglomerations in China, such as the Beijing-Tianjin-Hebei region, the Yangtze River Delta region, and the Fenwei Plain, are still suffering from severe air pollution (Zhang et al., 2019; Xiang et al., 2019; Haque et al., 2020; Zhao et al., 2020). Previous studies have shown that emission-reduction strategies, which are mainly based on the results of atmospheric chemistry simulations, play an important role in reducing pollutant concentrations and improving air quality (Zhang et al., 2016; Zhai et al., 2016). The emission inventory is an important basic data for atmospheric chemistry simulation, and its uncertainty will affect the accuracy of air pollution simulation, which in turn will affect the accuracy of pollution-control measures based on the model results (Huang et al., 2018). In order to improve the accuracy of atmospheric chemistry simulation and the feasibility of the pollution-control strategies, the emission data obtained by the “bottom-up” method needs to be inverted, which can be done through the atmospheric chemical variational assimilation system, to reduce the impact of emission uncertainty. GRAPES-CUACE is an atmospheric chemistry model system developed by the Chinese Academy of Meteorological

Sciences (CAMS)(Gong and Zhang, 2008; Zhou et al., 2008, 2012; Wang et al., 2010, 2015). GRAPES (Global/Regional Assimilation and PrEdiction System) is a numerical weather prediction system built by China Meteorological Administration (CMA), and it can be used as a global model (GRAPES-GFS) or as a regional mesoscale model (GRAPES-Meso) (Chen et al., 2008; Zhang and Shen, 2008). CUACE (CMA Unified Atmospheric Chemistry Environmental Forecasting System) is a unified atmospheric chemistry model constructed by CAMS to study both air quality forecasting and climate change(Gong and Zhang, 2008; Zhou et al., 2008, 2012). Using the meteorological fields provided by GRAPES-Meso, the GRAPES-CUACE model has been realized the online coupling of meteorology and chemistry (Gong and Zhang, 2008; Zhou et al., 2008, 2012; Wang et al., 2010, 2015). The GRAPES-CUACE model not only plays an important role in the scientific researches on air pollution in China (Gong and Zhang, 2008; Zhou et al., 2008, 2012; Wang et al., 2010, 2015, 2018), but also has been officially put into operation since 2014 at National Meteorological Center of CMA for providing guidance for air quality forecasting over China (Ke, 2019). Recently, An et al. (2016) constructed the aerosol adjoint module of GRAPES-CUACE model, which has been subsequently applied in tracking influential BC and PM2.5 source areas in north China (Zhai et al., 2018; Wang et al., 2018a, 2018b, 2019). However, these applications of GRAPES-CUACE aerosol adjoint model are still limited to sensitivity analysis, and the sensitivity information is not fully used to solve various optimization problems mentioned above. At the same time, considering the current severe pollution situation in mega urban agglomerations in China, more accurate emission data are urgently required to formulate reasonable and effective pollution-control strategies. In this study, we developed a new 4D-Var data assimilation system on the basis of the GRAPES-CUACE adjoint model, which was adapted for assimilating surface BC concentrations and optimizing its daily emissions in North China on July 4th 2016, when high BC concentrations observed in Beijing. ”

Section 2.1: Given this is a paper in GMD, the description of the model needs to be much more complete. Here is a brief list of immediate questions; the authors would

need to revise to address these and also provide a more detailed and complete description of the model in general. Reply: According to the reviewer's good instructions, we have provided a more detailed and complete description of GRAPES-CUACE atmospheric chemistry model and its adjoint model. For details, please see Section 2 in the revised manuscript.

What gas phase species are included? Does the model account for gas phase oxidation SO2 to SO4, or NOx to HNO3? Several recent papers have shown that NH3 plays an important role in governing aerosol concentrations in this region, given its role ammonium nitrate. However, it appears this model does not include NH3 or NH4, is that correct? How is missing this key species going to impact the results of this model? Another significant component of urban PM2.5 in the BTH region is secondary organic aerosol (SOA). Is any SOA included in this model, or is all the organic carbon assumed to be primary? If not, how does this bias the resulting calculations? How do the authors convert from mass of organic carbon to total mass for estimating PM2.5? Is the PM2.5 concentration estimated at dry or wet (i.e. including H2O in the mass) conditions, and if so, at what RH? Reply: Gas phase species in GRAPES-CUACE atmospheric chemistry model include SO2, NOx, NH3, CO, CO2, CH4 and non-methane volatile organic compounds (VOC). The GRAPES-CUACE model accounts for gas phase oxidation SO2 to SO4 and NOx to HNO3, and it also include NH3 and NH4. In GRAPES-CUACE model, six types of aerosols (sulfate, nitrate, sea salt, black carbon, organic carbon and siol dust) are formed in the aerosol module and they are segregated into 12 size bins with diameter ranging from 0.01 to 40.96 μm, while the ammonium is formed through the thermodynamic equilibrium module (ISORROPIA). Our research object is black carbon, so we didn't add the description of ammonium in the manuscript published on GMDD. According to the reviewer's good suggestions, we have added a more detailed model description this time. SOA in GRAPES-CUACE model is calculated through the aerosol and gas modules. Not all the organic carbon is assumed to be primary, and the organic carbon generated from gaseous VOC is also calculated in the gas module. The PM2.5 concentration estimated at dry conditions. We have added

a more complete description of the aerosol module, gas module and the thermodynamic equilibrium module in GRAPES-CUACE model in the part of Methodology as follow: “The atmospheric chemistry model CUACE mainly includes three modules: the aerosol module(module_ae_cam), the gaseous chemistry module(module_gas_radm) and the thermodynamic equilibrium module(module_isopia) (Gong and Zhang, 2008; Zhou et al., 2008, 2012; Wang et al., 2010, 2015). The interface program that connects CUACE and GRAPES-Meso is called aerosol_driver.F. This program transmits the meteorological fields calculated in GRAPES-Meso and the emission data processed as needed to each module of CUACE. The physical and chemical processes of 66 gas species and 7 aerosol species (sulfate, nitrate, sea salt, black carbon, organic carbon, siol dust and ammonium) in the atmosphere are comprehensively considered in the CUACE model (Wang et al., 2015). CUACE adopts CAM (Canadian Aerosol Module; Gong et al., 2003) and RADM II (the second-generation Regional Acid Deposition Model; Stockwell et al., 1990) as its aerosol module and gaseous chemistry module, respectively. CAM involves six types of aerosols: sulfate(SF), nitrate(NI), sea salt(SS), black carbon(BC), organic carbon(OC) and siol dust(SD), which are segregated into 12 size bins with diameter ranging from 0.01 to 40.96 μm according to the multiphase multicomponent aerosol particle size separation algorithm (Gong et al., 2003; Zhou et al.,2008, 2012; Wang et al., 2010, 2015). CAM also calculates the vertical diffusion trend of aerosol particles by solving the vertical diffusion equation. The core of CAM is the aerosol physical and chemical processes, including hygroscopic growth, coagulation, nucleation, condensation, dry deposition/sedimentation, below-cloud scavenging, and aerosol activation, which is coherently integrated with the gaseous chemistry in CUACE(Gong et al., 2003; Zhou et al.,2008, 2012; Wang et al., 2010, 2015). The gas chemistry provides the production rates of sulphate aerosols and secondary organic aerosols (SOA), and meanwhile generates an oxidation background for aqueous phase aerosol chemistry, in which sulphate transformation changes the distributions of SO2 in clouds (Zhou et al., 2012). Both nucleation and condensation are considered for sulphate aerosol formation depending on the atmospheric state after gaseous H2SO4

formed from the oxidation of sulphurous gases such as SO2, H2S and DMS(Zhou et al., 2012). Second organic aerosols as generated from gaseous precursors are partitioning among different bins through condensation using the same approach as the gaseous H2SO4 condensation to sulphate (Zhou et al., 2012). Given that the nitrates(NI) and ammonium(AM) formed through the gaseous oxidation are unstable and prone to further decomposition back to their precursors, CUACE adopts ISORROPIA to calculate the thermodynamic equilibrium between them and their gas precursors (West et al., 1998; Nenes et al., 1998a, 1998b; Yu et al., 2005; Zhou et al., 2012). ISORROPIA contains 15 equilibrium reactions, and the main species include gas phase: NH3, HNO3, HCL, H2O, liquid phase: NH+ 4, Na+, H+, Cl−,NO− 3, SO2− 4, HSO− 4, OH−, H2O, and solid phase: (NH4)2SO4, NH4HSO4, (NH4)3H(SO4)2, NH4NO3, NH4Cl, NaCl, NaNO3, NaHSO4, Na2SO4 (Nenes et al., 1998a). ”

There should be discussion of model evaluation, ie comparison against in situ measurements, either here or elsewhere. Reply: According to previous studies, the GRAPES-CUACE model can reasonably simulate the spatial distribution and temporal trend of PM2.5 and BC concentrations, and the correlation coefficient between the simulated concentration and the observed concentration of PM2.5 is about 0.5-0.9 (Wang et al., 2018a, 2018b), the and the correlation coefficient between the simulated concentration and the observed concentration of BC is about 0.6 (An et al., 2016). We also compared the simulated BC concentration in July 2016 against in situ measurements. The correlation coefficient is about 0.4 and the simulated BC concentration is relatively higher than the observed BC concentration. Since the priori emission used in this study was based on statistical data of anthropogenic emissions for 2007 (Cao et al., 2011). We were going to show the model evaluation with the priori and the optimized BC emission. However, we did not obtain the satisfactory results of the emission inversion within the specified time. Therefore, we did not write this part of the content into the revised manuscript.

134-138: This sounds like 4D-Var, unless the GRAPES-CUACES model itself is only

2D. Can the authors please clarify why they describe their system as 3D-Var? Reply: The GRAPES-CUACE model itself is 3D. Since the optimization was performed offline in our previous experiment, we thought that in a strict sense, this assimilation system cannot be called 4D-Var. In the revised manuscript, we have upgraded the code that was executed online and conformed to the description of 4D-Var.

140-142: Being “classic” or “simple” isn’t necessarily a good thing. In this case, the steepest decent method can be highly inefficient, requiring many more iterations to reach the minimum than other approaches. Given the wide availability of more advanced gradient-based minimization algorithms (such as quasi-newton variable metric methods like L-BGFS-B), it’s not clear why the authors chose the steepest descent method. Reply: In the previous experiment, the code of L-BFGS-B was not successfully debugged. So we used the steepest descent method instead. According to the reviewer’s good suggestions, we have re-debugged the L-BFGS-B code and successfully applied it to the assimilation system this time.

Section 2.5: Here is where the authors should describe in detail the following highly critical components of the inversion: gamma, B and R. What are their values? What assumptions were made in comping up with those values? What simulation experiments were considered to evaluate how robust the model inversion performance is for these values? etc. Reply: According to the reviewer’s valuable suggestions, we have described gamma, B, and R in detail as follows: “Based on Bayesian theory and the assumption of Gaussian error distributions (Rodgers, 2000), the cost function of the emission inversion is generally defined as follow: J(x)=1/2 ãĂŰγ(x-x_b)ãĂŮˆT Bˆ(-1) (x-x_b)+1/2 $\sum\_(i = 0)^p (F\_i(x) - y\_i)^T R^(-1)(F\_i(x) - y\_i)(9) where x, which we sought to optimize, generally represents the state vector of emis$ is the regularization parameter.” “According to Cao et al. (2011), the uncertainty of priori BC emissions used in this study is 76.2%. Therefore, we assigned the priori error covariance matrix (B) to be diagonal and the uncertainty to be 76.2% for BC emissions. Due to the lack of information to completely construct a physically representative B, the

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regularization parameter γ is introduced to balance the background and observation terms in the cost function. As described in Henze et al. (2009), an optimal value of γ can be found with the L-curve method (Hansen, 1998). Here, we followed this method, and obtained γ=0.01 through several emission inversions with a range of γ (10, 1, 0.1, 0.01, 0.001, 0.0001, 0.00001). " "The observation error covariance matrix (R), which is difficult to quantify, generally includes contributions from the measurement error, the representation error, and the forward model error (Henze et al., 2009; Zhang et al., 2016; Cao et al., 2018). And there is also a certain error in calculating the BC concentration based on the BC/PM2.5 ratio. To reflect the possibly large uncertainties of the observation, we assumed R to be diagonal and with error of 100%."

Section 2.6: Here the authors provide a few sentences about the data that is missing (ie, measurements of BC), but they provide very little information about the data that is actually used. Instead, they need to describe the measurements used in this study, addressing the following points: how many cites? where are they located (provide figure)? what species do they measure? at what RH are the masses recorded? what is the frequency of the observations? what instruments are used to make the measurements? Reply: According to the reviewer's good instructions, we have added a more detailed description of BC and PM2.5 measurements used in this study as follow: "The surface measurements of BC were collected from China Atmosphere Watch Network (CAWNET). The CAWNET was established by China Meteorological Administration to monitor the BC surface mass concentration over China in 2004, and had 54 monitoring stations in the summer of 2016. The monitoring of BC was conducted by an aethalometer (Model AE 31, Magee Scientific Co., USA), which uses a continuous optical gray scale measurement method to produce real-time BC data (Gong et al., 2019). In this study, we used the recommended mass absorption coefficient for the instrument at an 880-nm wavelength with 24-hour mean values of BC during July 1-31, 2016 at 5 representative stations of CAWNET in North China (Fig.S1). The surface PM2.5 concentrations were obtained from the public website of China Ministry of Ecology and Environment (MEE) (http://www.mee.gov.cn/). The network started to release

real-time hourly concentrations of SO2, NO2, CO, ozone (O3), PM2.5 and PM10 in 74 major Chinese cities since January 2013, which further increased to 338 cities in 2016. The PM2.5 data were collected by the TEOM1405-F monitor, which draws ambient air through a sample filter at constant flow rate, continuously weighing the filter and calculates the near real-time mass concentration of the collected particulate matter. We used hourly surface PM2.5 concentrations for July 1-31, 2016 at 48 cities in North China, including 12 cities in the Beijng-Tianjin-Hebei region (Fig.S1). Here, we have averaged PM2.5 concentrations at several monitoring sites in each city to represent a regional condition. To improve the performance of emission inversion, adequate observations are needed for constraining the model. Due to the limited BC monitoring sites in North China, we used the surface PM2.5 concentrations at 48 cities described above and the BC/PM2.5 ratio to obtain the hourly BC concentrations for July 1-31, 2016 at 48 cities in North China. The detailed calculation process can be found in the Supplement. ”

Figure S1. Simulation domain over North China. The bule stars denote locations of 5 BC surface monitoring sites from CAWNET. The details of the 5 sites in CAWNET are shown in Table S1. The dots denote locations of 48 PM2.5 surface monitoring sites from MEE with the city names labelled around. The red dots represent the 36 cities that are put into the assimilation system, and the green dots represent the 12 cities that are not put into the assimilation system and are used for validation of the effect of emission inversion. Table S1. Information of the 5 sites in CAWNET. Station Longitude Latitude Beijing 116.47°E 39.8°N Zhurihe 112.9 42.4 Yushe 112.98 37.07 Huimin 117.53 37.05 Zhengzhou 112.9 42.4

190-200: So the inversion is only performed for this brief period of a few days? While a good place to start, this seems very preliminary and only the first step of a study that needs to be completed prior to being published. Reply: The inversion can be performed for a longer period (i.e. one month). Since the parallel computing mode of the adjoint model and the assimilation system used in this study is still in the development stage,

the emission inversion can only be performed in a serial mode at present. It takes about 240 hours to execute one iteration of a 1-month simulation under the conditions of serial mode. In general, it takes 10-12 iterations to converge (Zhang et al., 2016). Thus, it will take at least 2400 hours (100 days) to converge if the emission inversion is performed for one month. Therefore, the emission inversion is only performed for one day, which is indeed very preliminary and only the first step of a study, as the reviewer pointed out. In the future, when the adjoint model and the assimilation system is successfully implemented in parallel mode, we will perform emission inversion for a longer period.

197: It is not clear to me what the authors mean by “inversion area” âAËŸT is this the ËĞ only region over which they are optimizing the emissions? Why would they artificially define such a region? The adjoint model should give them gradients with respect to each emission within their model domain, and any emission with a significant gradient should be optimized. Only adjusting emission in a pre-defined region will bias the results by over-adjusting these emissions. Reply: In the previous emission inversion, “inversion area” is the only region over which they are optimizing the emissions. We artificially defined such a region to reduce calculation and storage. We have realized that this is inappropriate. And we have redesigned the emission inversion, in which the gradients with respect to each emission within the model domain were included, and any emission with a significant gradient would be optimized. Thanks to the reviewer for helping us correct this problem.

205: Two major problems here: (1) Scientifically, this is a very bad idea; source inversion problems are ill-posed. The authors need to include some sort of prior constraint /regularization. The paper is recommended for rejection based on this alone. Note that if the penalty term is removed and R is assumed to be a uniform diagonal matrix, R itself has little to no impact on the inversion, which is also not good. Well formulated inversions are critically dependent upon R and B âAËŸT this needs to be taken further ËĞ into account. Reply: Thanks for pointing this out. We have realized that our

approach is inappropriate, and we have redesigned the emission inversion, in which the penalty term was not removed and R and B were taken into account. Please see section 3.1 in the revised manuscript or the response to problem (2) below.

(2) In terms of manuscript presentation, the authors have already defined the cost function, in a different form, using different notation âAËŸT they need to introduce one ËĞ and only one set of notation and definition for the the cost function in section 2, and then use that in section 3. Introducing one but using another is confusing. Reply: We apologize for the confusing description in the previous manuscript. We have rewritten the description of cost function. And there was only one cost function was defined this time. The description of the cost function is as follows: "3.1 Cost function Based on Bayesian theory and the assumption of Gaussian error distributions (Rodgers, 2000), the cost function of the emission inversion is generally defined as follow: J(x)=1/2 ãĂŰγ(x-x_b)ãĂŮˆT Bˆ(-1) (x-x_b)+1/2 $\sum\_(i = 0)$ Θ$(F_i(x) - y_i)$ØT R(−1)$(F_i(x) - y_i)(9) where x, which we sought to optimize, generally represents the state vector of emis$ is the regularization parameter. In this study, we followed the method in Henze et al. (2009), and defined x as the state vector of scaling factors of BC emissions: x=lnâĄą(s/s_b) (10) where s is the state vector of the daily gridded emissions of BC at three vertical levels (non-point source on the ground, middle elevation point source at 50m and high elevation point source at 120m), s_b is the priori estimate of s. Thus, the priori estimate of x (x_b) is equal to zero. According to Cao et al. (2011), the uncertainty of priori BC emissions used in this study is 76.2%. Therefore, we assigned the priori error covariance matrix (B) to be diagonal and the uncertainty to be 76.2% for BC emissions. Due to the lack of information to completely construct a physically representative B, the regularization parameter γ is introduced to balance the background and observation terms in the cost function. As described in Henze et al. (2009), an optimal value of γ can be found with the L-curve method (Hansen, 1998). Here, we followed this method, and obtained γ=0.01 through several emission inversions with a range of γ (10, 1, 0.1, 0.01, 0.001, 0.0001, 0.00001). "

220 - 225: This interpretation is not correct. For a multiplicative relationship of a*b*10, one cannot ascribe the factor of 10 as being related to b any more than a, since both a*(b*10) and (a*10)*b are equivalent. The introduction of these subjective rescaling of 10 or 100 is arbitrary and likely just compensation for some other deficiency in the inversion framework. The Bayesian approach for including prior information in the inversion via equation 6 is the best way to incorporate the prior into the inversion framework; the authors should stick with that approach. Reply: Thanks for pointing this out. We have realized that the interpretation is not correct. And we insisted on Bayesian approach in the revised manuscript, as mentioned above in the response to problem (2).

4.3: The authors use a comparison of the posterior model to the observations used during the inversion as a means of validation. This is a rather weak test, as by definition if the cost function has reduced, then the model is going to do a better job of matching these observations. Instead, the authors should test against an independent set of observations (not used during the inversion), or consider methods for calculating the posterior error in the emissions themselves (comparison to other studies, estimation using numerical techniques from the inversion, etc). Reply: According to the reviewer's valuable suggestions, we were going to validate the inversion effect using the observations that were not used during the inversion. And we have addressed it in section 3.4 in the revised manuscript. We proposed that "The hourly BC concentrations at 36 cities during this time interval are adapted for the emission inversion, and the BC concentrations of the remaining 12 cities are used for validation of the inversion effect." In future work, we will also compare our results to other studies.

Code: I checked both web sites provided for downloading the GRAPES-CUACE model and did not see code download at either location. The urls provided are generic links for large institutions. This does not meet the minimum standard of code availability for GMD. Reply: We are sorry for not being able to download the GRAPES-CUACE code. Since the code of the GRAPES-CUACE model is not open source, and we are not the developers of the GRAPES-CUACE model, we have no right to make the code

public. The websites we provided are the official websites of the Numerical Weather Prediction Center of Chinese Meteorology Administration and the Chinese Academy of Meteorological Sciences. Only through official registration and authorization can the code of the GRAPES-CUACE model be obtained.

I also reviewed the code provided the supplemental. I do not see how this code accomplishes the steps described in the paper. I don't see where the information for the adjoint is input into the optimization routine, where the updated scaling factors are calculated, and where these are provided to the forward model to run, iteratively. The optimization seems to be running entirely offline, which would be consistent with 3D-Var; however the process described the manuscript and the accompanying flow chart (fig 3) shows a 4D-Var system. In general, the code is not well documented nor explained, which again is antithetical to the purpose of publishing in GMD. It also appears to allow for the cost function to have "converged" if it is increasing, rather than decreasing, which is odd. Shouldn't J(k+2) always be less than J(k+1)? Reply: As the reviewer understood, the optimization was performed entirely offline in our previous experiment. In the revised manuscript, we have adopted the L-BFGS-B method and upgraded the code, which was executed online and included where the information for the adjoint is input into the optimization routine, where the updated scaling factors are calculated, and where these are provided to the forward model to run, iteratively. We have also realized that it's not correct to allow for the cost function to have "converged" if it is increasing, rather than decreasing. During our testing of the new code, the cost function is always reduced (J(k+2) is always less than J(k+1)).

- The grammar and writing needs a lot of work throughout, pretty much every sentence. It goes beyond my responsibility to edit in this detail, but the authors will need to hire editing services or engage additional co-authors to help with this in their next submissions. Reply: We apologize for the typos and grammatical issues underlined by the reviewer. We have carefully revised the manuscript to improve this point specifically.

Other: Throughout: Use the phrase a priori / a posteriori OR prior / posterior consis-

tently throughout, and do not mix-and-match. Reply: Thanks for pointing this out. We used prior/posterior consistently throughout this time.

23 - 25: rewrite, not clear Reply: The expression of these sentences is inappropriate, so we deleted them.

42: What is meant by “reverse optimization”? Reply: We apologize for the inappropriate expression. What we wanted to express is the optimization of emission. We have corrected it in the revised manuscript.

82: I don’t think the nationality of the people who developed the model is scientifically relevant. Reply: Yes, the nationality of the people who developed the model is not scientifically relevant. We apologize for this expression. And we will never again appear similar inappropriate expressions.

128: Can the authors be more specific here? What was checked, and how accurate did they find the performance to be? Reply: The tangent linear model (TLM) and the adjoint code of the GRAPES-CUACE model were checked. According to the reviewer’s valuable suggestions, we have added more specific description of the adjoint validation as follow: “2.2.2 GRAPES-CUACE aerosol adjoint The GRAPES-CUACE aerosol adjoint model was constructed by An et al. (2016) based on the adjoint theory (Ye and Shen, 2006; Liu, 2005) and the CUACE aerosol module, which mainly includes the adjoint of physical and chemical processes and flux calculation processes of six types of aerosols (SF, NI, SS, BC, OC and SD) in CAM module, the adjoint of interface programs that connect GRAPES-Meso and CUACE, and the adjoint of aerosol transport processes. As described in An et al. (2016), after the construction of the adjoint model is completed, its accuracy must be verified to confirm its reliability. Since the adjoint model is built on the basis of the TLM, the validity of the TLM must be ensured before the accuracy of the adjoint model is tested. The verification formula of tangent linear codes can be expressed as: Index=limâŤň(δX→0)âĄąãĂŰ(F(X+δX)-F(X))/(δXFˆ’(X))=1.0ãĂů, (6) where the denominator is the TLM output, and the numerator is the

difference between the output value of the original model with input X+δX and input X. It is necessary to decrease the value of δX by an equal ratio and repeat the calculation of the above formula. If the result approaches 1.0, the tangent linear codes are correct. It was verified that all input variables in the model, such as the concentration value of pollutants (xrow) and the particle's wet radius (rhop), have passed the TLM test. The adjoint codes can be validated on the basis of the correct tangent linear codes. The adjoint codes and the tangent linear codes need to satisfy Eq.(2) for all possible combinations of X and Y. In Eq.(2), L and LâĹŮ represent the tangent linear process and the adjoint process, respectively. To simplify the testing process, the adjoint input is the tangent linear output: Y = L(X). Thus, Eq.(5) can be expressed as: (âĹĞF·dX,âĹĞF·dX)=(dX,âĹĞˆT F(âĹĞF·dX)). (7) By substituting dX into the tangent linear codes, the output value âĹĞF·dX can be obtained and the left part of the equation can be computed. Then, taking âĹĞF·dX as the input of the adjoint codes, the adjoint output âĹĞˆT F(âĹĞF·dX) can be obtained and the right part of the equation can be calculated. On condition that the left and right sides of EQ.(7) are equal within the range of machine errors, the constructed adjoint model is validated. It was verified that all input variables in the model have passed the adjoint test. Taking the pollutant concentration variable (xrow) as an example, both sides of EQ.(7) produce values with 14 identical significant digits or more. This result is within the range of machine errors, so the values of the left and the right sides are considered equal. Thus, the pollutant concentration variable (xrow) has passed the adjoint test."

150: It is more correct to say that the cost function is based on the Bayesian method. The adjoint is just a tool used for finding the minimum of this cost function. Strictly speaking, there is no such thing as an "adjoint inversion", since the adjoint is just one piece of the overall inversion framework. I know that people may say that, casually, but in scientific manuscripts we should be more careful. Bayesian method leads to the cost function. One approach to minimizing the cost function is 4D-Var. An adjoint is just a part of the numerical tools we often use as part of 4D-Var (keep in mind it is possible to do 4D-Var without an adjoint...). Reply: Thanks for pointing this out. We

have revised the expression as follow: “Based on Bayesian theory and the assumption of Gaussian error distributions (Rodgers, 2000), the cost function of the emission inversion is generally defined as follow: J(x)=1/2 ãĂŰγ(x-x_b)ãĂŮˆT Bˆ(-1) (x-x_b)+1/2 $\sum\_(i=0)$ (F_i(x) − y_i)ĘTR(−1)(F_i(x) − y_i)(9)″

161: Technically FËĘT is the transpose of the model Jacobian. In contrast, the adjoint model is what is used to solve the product of FËĘT and a vector. The vector here would be the adjoint forcing, RËĘ(-1)(H(x) - y) Reply: Thanks for pointing this out. In the future, we will pay attention to using correct and rigorous expressions.

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