Referee #1

Received and published: 12 January 2016

This paper presents the treatment of aerosol background error covariance with balance constraints. Overall, the paper is well written. The presented result shows that the method would improve the chemical data assimilation performance.

We thank Reviewer #1 for thoroughly reviewing the manuscript, valuable comments and constructive suggestions. We have carefully addressed all Reviewer's comments and suggestions. We also respond point by point to the reviewer's comments as listed below.

One major concern is that the numerical experiments were only based on a 24-hour forecasting. Since the atmospheric chemistry and meteorological conditions vary day to day. It is highly suggested that the authors extend the experiments to a longer time period. The test period is coincident with CalNex field campaign. So it is not difficult to find more observations for such testing.

The period of the effect of data assimilation is generally less than 24 hours (Fig. 12). A longer time forecast of the DA experiment should be very close to the experiment without DA. To demonstrate the robustness of our DA system, we conducted nine cases with a group of 24-h forecasts for each case. For the flight events are discontinuous, we ran these cases with different initial time according the flight processes. The details of these nine cases are in Table 2 (Page 19) and Figure 8 (Page 21). And the results are showed in Figure 11 (Page 26) and Figure 12 (Page 28).

Cross-correlations can be between different species/bins or different grid points. The authors often use "cross-correlation" without specifying what they mean. It is helpful to be unambiguous. For instance, in abstract (line 7 on Page 10054), "cross-correlation" probably refers to the correlation between different species.

Thanks, the specification for cross-correlation has been revised in this sentence and some other sentences (line 40, line 123, line 141, line 191, line 268, line 269 and line 580).

The description of model configuration is lacking. Although readers are referred to Li et al. (2013) for details, some basic information should be provided directly. For instance, the mapping projection used for the horizontal coordinates and the extensions of the vertical levels are better given in the paper.

We have added some description of the model configuration in the revised manuscript (lines 273-277).

The calculation of the cross-correlation of emission species is not clear. Is it based on 15 May-14 June, 2010 emissions over domain d03?

The emission species are referred to those RADM2 species that produced by NEI’05 data. We calculated the correlation between any two horizontal fields of emission species over domain d03. It is indicated at line 294.

10054, line 14, "are more coincident" -> have better agreement

Thanks, the sentence has been revised (line 47-49).

10054, line 23, "meteorology-chemistry models" -> Chemical transport models

The "meteorology-chemistry models" has been revised as “Chemical transport models” (lines 62).

10055, line 4, "difficult dealt due to ...": Remove "dealt".

The “dealt” has been removed.

10055, line 26, "balance analysis fields": balanced analysis fields?

Corrected.

10056, line 16: PM2.5 is part of PM10 and PM1 is part of PM2.5. So they do not represent different size bins.

This sentence has been revised, and we cited two new papers about the relationship of PM2.5 and PM10-2.5 (lines 109-111).

10056, line 19: The spread of observation impact is not necessarily “enhanced”.

Corrected. We have changed to “produce more balanced initial fields” (line 109).

10057, line 4: It is not clear what “the species that are not ADJACENT” means here.

“that are not ADJACENT” means that are not the connecting. This sentence has been revised (line 121-122).

10057, line 12, “.. has been ESTIMATED ...”: Developed or applied?

The “estimated” has been changed to “developed’’ in the revised manuscript.

10058, Eq(1): It is better to have the LHS written as J(x) and x should be in bold font.

“$J(\delta x)$” has been changed to “$J(\boldsymbol{x})$”, and $\boldsymbol{x}$ is in bold font in the revised manuscript.

10059, line 2, “d = y - Hx ”: d= y-H xˆb

Corrected.

10059, lines 6-7: This seems to neglect the fact that there are multiple variables at each grid point.

Corrected. The sentence has been change to: For a high-resolution model, the number of vector $\boldsymbol{x}^{\boldsymbol{b}}$ is on the order of 10^7. Therefore, the number of elements in **B** is approximately 10^{14} (line 174).

10059, line 18, “which represent ...”: Separate the run-together sentence. They represent the correlation among pairs of grid points for one species.
Thanks. The sentence has been separated (lines 185-186).

10063, line 16, “cross-correlations between emissions”: Change to “cross-correlations of emission species”, to be consistent with the title of Section 3.2.
The sentence has been revised (Line 265).

10063, line 19, “the cross-correlations of aerosol emissions from ...” -> the cross-correlations of aerosol emission species from ...
The sentence has been revised (Line 268).

10064, line 3, “...that is coupled to aerosol and chemistry domains” -> ...that is coupled to aerosol and chemistry models
The sentence has been revised (Line 272).

10064, lines 16-17, “Emission files are .... of the aerosol forecasts”: What does "a primary factor for the distribution of the aerosol forecasts” mean? In addition, it is a run-together sentence that needs to be rewritten.
“A primary factor” means the emission file. This sentence has been revised as: The emission files are necessary for running the WRF/Chem model. It is an important factor for the distribution of the aerosol forecasts. (lines 290-291)

10065, line 5: “With the exception of the auto-correlation in the diagonal line” is redundant.
The sentence has been removed.

10066, line 25: Please spell out “DA” as “data assimilation” since DA is not previously defined yet.
Corrected.

10067, line 3: Figure 2 -> Figure 4.
Corrected.

10067, line 4: Fig. 2a -> Fig. 4a.
Corrected.

10067, line 8: Fig. 2b -> Fig. 4b.

Corrected.

10067, line 8, “all standard deviations significantly decrease”: The decrease of NO3 is not significant.

The sentence has been change to: “all standard deviations decrease in different degrees” (Line 367).

10069, line 8: There are many other flights available. Why was this flight chosen over all the others? More description on the flight observations is needed as well.

We have added all flight events that performed during May 15 to 00UTC of June 14, 2010. We chose the case of June 3 for the aircraft observations are enough and the aircraft tracks around Los Angeles, the center of model domain. For the other cases, there are not enough aircraft observations during the assimilation time windows (±1.5 hour of initial time), or the flight tracks are relative few and not around Los Angeles (Fig. 8). But, to demonstrate the robustness of our DA system, we run all cases and calculate the average improvements.

10069, line 25: WRf/Chem -> WRF-Chem. Note that the WRF/Chem is better changed to WRF-Chem in the entire paper.

All “WRF-Chem” have been change to “WRF/Chem”.

10071, line 17: Figure 11 does NOT show “scatter plots”.

Corrected, thank you.

10071, line 19: Fig.1a -> Fig. 11a

Corrected, thank you.

10072, lines 9-10: It is not true that the data assimilation has the hypothesis of the independent control variables. The independence of control variables merely helps to simplify the background error covariance matrix.

We agree. This sentence has been removed.

10077, Table 1: Please add the name of the species to the table.

Corrected, thank you (Page 33).

10079, Figure 2: Why aren’t the cells identical in shape? It applies to Figure 3 too.

Figure 2 and Figure 3 have been plotted in the same shape.

10081, Figure 4, “Same as Fig. 3”: Figure 4 is quite different from Figure 3.
Corrected, thank you (line 373-374).

Referee #2

This paper discusses implementation of cross-correlations between aerosol variables in a variational data assimilation (DA) system via balance constraint. The authors describe their methodology and then apply their new developments for a single 24-hr forecast over Southern California. Results suggest that incorporating cross-correlations within the DA system was beneficial, especially for 3- to 18-hr forecasts.

This paper is generally interesting and good, although there are some shortcomings that I believe should be addressed before publication. My biggest concerns regard that only one forecast was produced and lack of discussion about other methods of dealing with cross-correlations for aerosols, such as ensemble-based DA methods. Additionally, there are many areas of text that I believe require some clarification.

Thank you very much for your careful review and constructive suggestions. Please find below our detailed responses to all questions and comments.

Bigger comments, questions, and concerns

1. I appreciate that you actually implemented your developments in a DA system to see the real-world impacts. However, you only showed results from one forecast, which does not give much confidence regarding the generality or strength of the results. If possible, I strongly urge you to add more cases. I know that adding more cases requires more work, but doing so would not add much to the length of the paper and would make the conclusions much more robust.

We agree that single one case is not convincing to show the capability of our DA system, though the major purpose is to develop the DA system with the cross-correlation process. We have added more cases in the revised manuscript, all nine flight cases from May 15 to June 14, 2010, are applied to DA experiments.

The details of these nine cases are in Table 2 and Figure 8. The results are showed in Figure 11 and Figure 12. The averaged improvement of DA for these nine cases is lower than that for the case on June 3, 2010. The main reasons are that the flight tracks are relative fewer in some cases, or the flight tracks are not around Los Angeles. Another reason is that the initial fields of Control experiment are consistent with observations, especially for the initial time at 00 UTC and 18 UTC. In the case that limited improvements were obtained at the initial fields, the improvements of subsequent forecasts are also low.

2. In my opinion, you neglected to discuss another (and easier) method of dealing with cross-correlations between aerosol species: ensemble-based DA methods (such

as the ensemble Kalman filter) that naturally handle cross-correlations. Thus, I strongly believe you should mention ensemble DA methods in the introduction, and you should cite and briefly discuss Pagowski and Grell (2012) and Schwartz et al. (2014), who assimilated aerosol observations, including PM2.5, with ensemble-based DA methods. There are other references that have also assimilated aerosol observations with ensemble DA, but I believe those two are the most relevant, and without this material regarding ensemble DA, I believe your work is not placed within its proper context.

Thanks. These two papers have been cited, and some discussions about ensemble DA methods are added in the revised manuscript (lines 109-111).

3. In light of the above comment, I believe your title should be more specific, and I suggest adding the word "variational" before "data assimilation".

We agree. The title has been revised.

4. You left out a few important details about the DA system. For example, what DA system were you using? Was it GSI or some other system? Please briefly explain somewhere in the text. Additionally, for your 24-hr forecast you described in section 5, what was the background for DA? Finally, please briefly state the observation errors that you used.

Firstly, this DA system is not GSI or some other widely used systems. It was developed by Li et al. (2013) for the MOSAIC scheme of WRF/Chem model. A simple description about the DA system was added in Section 1 (line 129-130).

Secondly, the backgrounds for DA are the forecasting results from the previous runs without DA. These previous forecasting results have been obtained when we run the model for the BEC statistics. We added the description of the background in Section 5.1 (lines 471-473).

Thirdly, we assume that the observation error is the half of background errors. And a vertical profile of observation errors was applied, resulted from the average of background errors of every level. We think it is an enough large error, even the representativeness error is considered. Since the purpose of this manuscript is to demonstrate the signification of balance constrains in the 3DVAR system, the observation error has an insignificant impact on the analysis of balance constrains. We added the description of the observation error in Section 5.1 (lines 473-475).

5. I believe some aspects regarding Eqs. (6-13) need clarification.

a) Page 8, line 9: Please clarify what you mean by "first variable".

The "first variable" means this variable is fixed. There is not unbalanced component for this variable that is similar to the variable of the vorticity in the DA system of ECMWF (Derber and Bouttier, 1999). But, we did not find the name of "first variable" in other relevant literatures. Thus, we have removed this name in the revised manuscript.

b) Page 8, Eq. (7): Please fill-in the upper triangle of **K**. Are all upper-triangle elements zero?

Yes, all upper-triangle elements are zero. We have filled in the matrix.

c) Page 8, line 19: Please clarify what you mean by “a one regression coefficient.”

It is a mistake. It should be “a regression coefficient”. We have revised this sentence.

d) Some more details about how you compute *ρij* would be beneficial.

The $\boldsymbol{\rho}_{\mathbf{ij}}$ is the statistical regression coefficients between the variables i and $\boldsymbol{j}$**.** For example $\boldsymbol{\rho}_{\mathbf{12}}$ is the regression coefficient between δEC and δOC. Here, δEC and δOC are estimated from the forecast differences of 24 h and 48 h forecasts of one month (May 15 to June 14, 2010), that is $\boldsymbol{\delta EC = EC^{24} - EC^{48}}$, $\boldsymbol{\delta OC = OC^{24} - OC^{48}}$. Similar to the calculation of the BEC, $\boldsymbol{\delta EC}$ and $\boldsymbol{\delta OC}$ are also estimated by this forecast difference to represent the difference between real state and forecasts. Using the forecast difference, we have 30 pairs δEC and δOC for each grid. Then, we can estimate a regression equation and obtain the regression coefficient $\boldsymbol{\rho}_{\mathbf{21}}$. Since the $\boldsymbol{\rho}_{\mathbf{21}}$ of each grid are close, we use the data of $\boldsymbol{\delta EC}$ and $\boldsymbol{\delta OC}$ at all grids to estimate a regression equation and obtain a regression coefficient ρ_{21}. This ρ_{21} should be more robust. Figure 1 shows the scatter plots of $\boldsymbol{\delta EC}$ and $\boldsymbol{\delta OC}$ for all grids. The size of $\boldsymbol{\delta EC}$ or $\boldsymbol{\delta OC}$ is $(\mathrm{N} \times 30)$, where N is the number of model grid points, and 30 represents 30 days. From this scatter plots, we can obtain the regression equation:

$$\widehat{\boldsymbol{\delta OC}} = 0.9 \times \boldsymbol{\delta EC}. \qquad (1)$$

$\widehat{\boldsymbol{\delta OC}}$ is the predict of $\boldsymbol{\delta OC,}$ that is $\boldsymbol{\delta OC_b}$. The residuals are $\boldsymbol{\delta OC_u}$**.** In this equation, the intercept is neglected, since δOC and δEC are forecast differences that can be considered to be zero mean values. Using $\boldsymbol{\delta NO_3}$ $\boldsymbol{\delta EC}$ and $\boldsymbol{\delta OC_u}$, we can estimate the regression equation to predict $\boldsymbol{\delta NO_{3b}}$, and obtain $\mathrm{\rho}_{31}$ and $\mathrm{\rho}_{32}$.

$$\boldsymbol{\delta NO_{3b}} = \widehat{\boldsymbol{\delta NO_3}} = \mathrm{\rho}_{31}\boldsymbol{\delta EC} + \mathrm{\rho}_{32}\boldsymbol{\delta OC_u}. \qquad (2)$$

Then, the other regression equations and regression coefficient can be obtain step by step. Some more detail about the calculation of $\boldsymbol{\rho}_{\mathbf{ij}}$ was added in the revised manuscript (lines 327-331).

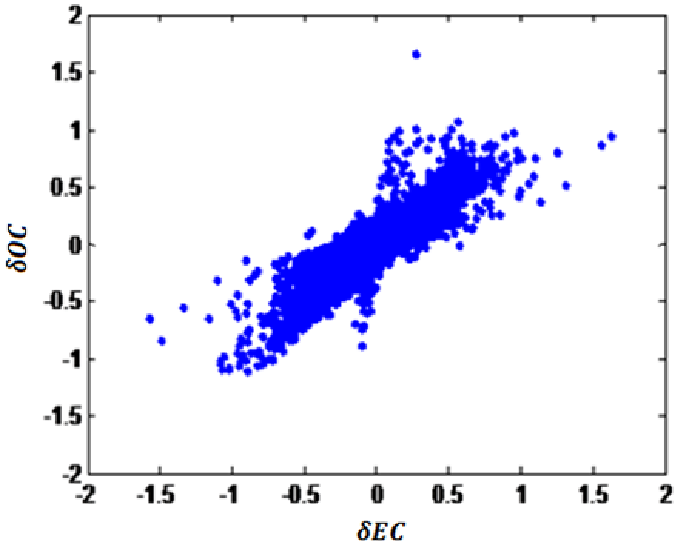

Figure 1 Scatter plots of $\boldsymbol{\delta EC}$ and $\boldsymbol{\delta OC}$

e) Additionally, I think it would be nice if you provided some details on how to interpret ρij to bolster the discussion on page 14.

We agree. Some details of the calculation of ρ_{ij} are added in the revised manuscript (lines 327-331).

f) What would happen if the regression was not "based" on EC? In other words, what would happen if you listed the control vector species in reverse [such that OTR was in the first row on the LHS of Eq. (7) and EC was in the last row]? You mention some of this on page 14 lines 6-8, but I believe a clear description about the "order" or "first and second variables" would be greatly beneficial. You also mention using OTR as the "last variable" (page 14, line 19), but the rationale for this choice is not obvious to me. Please clarify.

We think it is difficult to clarify this question which is beyond the scope of current study. We set this order of species mainly due to the following two reasons. First, the correlation of EC and OC is the highest. Second, OTR is correlative with all other variables. The purpose of the balance constrain is to obtain as independent variables as possible. So, EC and OC should be the first two for their high correlation. If we set the other variable such as OTR as the first order, and OC as the second order, the coefficient of determination of the regression equation of OC will be less, compared with the coefficient of determination of the regression equation with EC as the first order. It will increase the correlation of OC_u with the other variables. Similarly, since OTR includes many species that are correlative with former variables, the coefficient of determination of the regression equations of OTR will be largest using all former variables as factors, and then obtain the more independent OTR_u. To investigate the impacts by using different orders, more tests need to be conducted which we may address in later studies.

In addition, we can refer other DA system to understand this question. In the formulation of DA of ECMWF (Derber and Bouttier, 1999), The balance operator **K** matrix which transforms $[\zeta, \eta_{\mathrm{u}}, (T, P_s)_{\mathrm{u}}, q\ ]$ into $[\zeta, \eta, (T, P_{\mathrm{s}}),\ q\ ]$. The first variable is the vorticity ζ The balanced part of the divergence η and the temperature and surface pressure (T, P_{s}) are given by the equations:

$$\eta_{\mathrm{b}} = \boldsymbol{M}\zeta, \tag{3}$$

$$(T, P_{\mathrm{s}})_{\mathrm{b}} = \boldsymbol{N}\zeta + P\eta_{\mathrm{u}}. \tag{4}$$

Then, the **K** matrix becomes:

$$\boldsymbol{K} = \begin{bmatrix} \boldsymbol{I} & \boldsymbol{0} & \boldsymbol{0} & \boldsymbol{0} \\ \boldsymbol{M} & \boldsymbol{I} & \boldsymbol{0} & \boldsymbol{0} \\ \boldsymbol{N} & \boldsymbol{P} & \boldsymbol{I} & \boldsymbol{0} \\ \boldsymbol{0} & \boldsymbol{0} & \boldsymbol{0} & \boldsymbol{I} \end{bmatrix}. \tag{5}$$

In this DA system of ECMWF, the ζ and η are the first two variables. We think the reason is that they are relative high correlative. The (T, P_{s}) is the third variable, since it is correlative with the former variables. The q is the last variable that is not correlative with the other variables. Unfortunately, Derber and Bouttier (1999) did not explain why they set this order of variables. We explain it from our thought.

In another reference about the study of balance constraints for GSI system (Chen et al., 2013), the order of control variables is the stream function (ψ), the unbalanced part of the velocity potential (χ_{u}), the unbalanced part of temperature (T_{u}), the unbalanced part of surface pressure (ps_{u}), and the relative humidity (rh_{u}). Here, rh_{u} is the last order, and its regression equation uses all former variables as factors.

g) Page 9, line 1: I feel like the word “deduced” to describe ρ21 is inaccurate. How exactly are you obtaining ρij?

The “deduced” has been changed to “obtained” (line 362). Please see the response of d) for the process of obtaining ρij.

h) Page 9, lines 3-10: I found **ε** confusing and also unnecessary. By definition, **ε** = δOCu, so why not just use δOCu directly in place of **ε**? Thus, I suggest removing all instances of **ε**.

We wrote Eq. (8) and reserve residual ε because it is a normal format for a regression equation. This may be easier to understand for the reader. And we revised the Eq. (9) and its explanation to understand easily the calculation of $\boldsymbol{\delta OC_b}$ and ε (line 215).

i) Page 9, Eq. (11). I believe you’re missing “δ” on EC and OCu.

Corrected, thank you.

Smaller comments, questions, and concerns

1. Page 2, line 14: Please clarify what you mean by “coincident”.

We mean the PM2.5 concentrations of the experiment with balance constraints are more consistent with the observed concentrations. The sentence has been removed.

2. Page 2, line 17: Please omit the word “significant” because you did not perform any statistical significance testing, and you only showed results from one forecast.

Thanks, the sentence has been revised.

3. Page 2, line 21: Again, omit “significantly”.

Thanks, the sentence has been revised.

4. Page 2, line 26: Technically, the observation errors also determine the analysis increments.

Thanks, the observation error has been added in the revised manuscript (line 65).

5. Page 3, line 5: Most models now have a state size O(10^7). Suggest modifying.

Thanks, the sentence has been revised (lines 69).

6. Page 3, lines 3-8: Note that with ensemble DA methods, these issues are not as difficult to deal with.

Thanks, we add the qualifier of “variational data assimilation system” (line 99).

7. Page 3, line 12: Please define in words what you mean by PM2.5.

Thanks, the definition of PM2.5 has been added in the revised manuscript (lines 105).

8. Page 3, line 13: Suggest spelling out GSI and adding a reference.

We have spelled out GSI in the revised manuscript (line 77).

9. Page 4, lines 9-11: Do these assumptions only apply to variational approaches?

Yes. We add the qualifier of “variational” in the revised manuscript (line 99).

10. Page 4, line 20: This might be a good place to mention Pagowski and Grell (2012) and Schwartz et al. (2014).

Thanks, we have cited these two references at lines of 109-110.

11. Page 5, line 1: Please spell out “AOD”.

Thanks, the sentence has been revised (line 119).

12. Page 5, line 4: Please clarify what you mean by “not adjacent”.

"that are not ADJACENT" means that are not the connecting. This sentence has been revised (line 121-122).

13. Page 5, line 10: Please clarify what you mean by "eight/four".
The MOSAIC scheme offers flexibility in specifying the number of size bins, four or eight bins are commonly used. Four bins used are located between 0.039–0.1 µm, 0.1–1.0 µm, 1.0–2.5 µm, and 2.5–10 µm. Some introductions of "eight/four" size bins have been added in the revised manuscript (lines 127-128).
14. Page 5, line 12: Suggest "developed" rather than "estimated".
Thanks, the sentence has been revised.

15. Page 6, Eq. (1): It should be J(x) not J(δx).
Thanks, the sentence has been revised (line 159).

16. Page 6, lines 20-25 and Eq. (2): You've ignored non-linear H and its linearization about the background to derive the linear H. In Eq. (1), H is nonlinear, but in Eq. (2) it's linear, because you've linearized H about xb. Please be more precise.
Thanks. In this paper, the observation variables are the species concentration and total PM2.5 concentration. They are really linear relationship with the state variables. Anyway, we have added the assumption of linear in the revised manuscript (line 163).

17. Page 7, line 2: In the expression for the innovation, here H should be nonlinear (H).
Since the relationship between observation variables and state variables is linear. We do not emphasize the nonlinear H.

18. Page 7, line 7: Again, it should probably be 10^7 rather than 10^6. Also, 10^{12} should probably be 10^{14}.
Thanks, the sentence has been revised (lines 174).

19. Page 7, line 14: Please clarify what you mean by "is commonly simplified with vertical levels."
The standard deviation matrix (D) is a diagonal matrix with the size of $(N \times m)^2$, that is, each species at each grid has a value of standard deviation. But, to reduce the computational cost, we use the average value of standard deviations that are at the same vertical level. Though the size of **D** is fixed, the number of parameters of standard deviations reduces in the DA system. We have added some introduction in the revised manuscript (lines 180-181).

20. Page 10, lines 8-9: It was unclear to me how you got Eq. (17) from Eq. (6).

Please add some steps or clarify.

According the definition of the BEC,

$$\mathbf{B} = \langle(\boldsymbol{\delta x})(\boldsymbol{\delta x}^{\mathbf{T}})\rangle.$$

Using Eq. (6),

$$\begin{aligned}\mathbf{B} &= \langle(\mathbf{K}\boldsymbol{\delta x}_{\mathbf{u}})(\mathbf{K}\boldsymbol{\delta x}_{\mathbf{u}})^{\mathbf{T}}\rangle \\ &=\langle(\mathbf{K}\boldsymbol{\delta x}_{\mathbf{u}})(\boldsymbol{\delta x}_{\mathbf{u}}{}^{\mathbf{T}}\mathbf{K}^{\mathbf{T}})\rangle \\ &= \mathbf{K}\mathbf{B}_{\mathbf{u}}\mathbf{K}^{\mathbf{T}}\end{aligned}$$

Some explains have been added in the revised manuscript (lines 237-242).

21. Page 11, line 1: In Eq. (20), it appears you used $\delta x = B^{1/2}\delta z$. Thus, I believe line 1 on page 11 should read $\delta z = B^{-1/2}\delta x$ (note the negative sign on the exponent of B). Please double-check.

Corrected, thank you.

22. Page 12, line 5: Should be “horizontal grid spacing” not “resolution”…they mean different things.

Thanks, the sentence has been revised (lines 274).

23. Page 12, lines 10-12: Please clarify what you mean by “former forecast”. Additionally, where do the initial meteorological conditions come from? Are these also from NARR?

The initial meteorological condition is from NARR. For a 48-hour or 24-hour forecast running, we update the initial meteorological condition using the reanalysis NARR data. But for the initial aerosol condition, since there are not reanalysis data, we use the forecast condition from former forecast as the initial condition. The explaination has been added in the revised manuscript (line 281-283).

24. Page 12, line 26 and page 13, line 2: I wonder if you might want to rename“E_ORG” to “E_OC” and “E_PM25” to “E_OTR” to be consistent with the nomenclature of the control variables. If so, please also change on the relevant figure (Fig. 2) and elsewhere in the text.

Since the emission variables are different with the model control variables. The former include many aerosol precursors such as E_SO2, E_NO2. These aerosol precursors can transform into aerosol through chemical process. Thus, the emission variable E_SO4 or E_NO3 are not completely corresponding to the control variables. We use the name of emission variables, consistent with the name in user’s guide of WRF/Chem.

25. Page 13, line 5: “With the exception” is misleading and suggests that the diagonal correlations will be < 0.5. Please modify.

We have modified the corresponding sentence in the revised manuscript (Line 305).

26. Page 13, line 9: Suggest “high” rather than “close”.
Corrected.

27. Page 14, line 1: I believe it should be Eqs. (6-13) rather than Eqs. (6-12).
Corrected.

28. Page 14, line 2: I believe Eq. (7) is more correct than Eq. (6).
Corrected.

29. Page 14, lines 9-19: Should the control variables here have subscripts “u”? I’m not sure. Please double-check.
We have double-checked.

30. Page 14: Just a comment—I really like Fig. 3.
Thanks.

31. Page 15, line 2: Suggest “obtained” rather than “performed”.
Corrected.

32. Page 15, lines 2, 3, and 8: In all these locations, it should be Fig. 4, not Fig. 2.
Corrected, thanks.

33. Page 15, lines 10-11: I believe OTR and NO3 should be OTRu and NO3u, respectively.
Corrected.

34. Page 15, lines 10-11: Please clarify with what the “decreases” are with respect to.
Thanks, we have modified the sentence (Line 369)

35. Page 15, line 17: I believe it should be Eq. (22), not Eq. (21).
Corrected.

36. Page 15, lines 18-25: Please explain how you get the horizontal correlation scale (Ls) from Fig. 5. Is Ls defined as an e-folding distance? Overall, I was a bit confused by your description of Ls —please clarify.
We assume that the decline curve of horizontal correlations is according to the Gaussian function (Fig. 5). Then the intersection of the decline curve and the line of $e^{-\frac{1}{2}}$ (≈ 0.61) can be approximately as the value of horizontal correlation scale. The introduction has been added in the revised manuscript (lines 384-386).

37. Page 15, line 27: I believe OC, NO3, SO4, and OTR should have subscript “u”.

Corrected.

38. Page 16, line 1: Please clarify what you mean by “common factors in regression equations”.

The common factors mean EC, $\mathrm{OC_u}$, and $\mathrm{NO_{3u}}$. For example, EC is used four times in the regression Eqs. (6-13), $\mathrm{OC_u}$ is used three times, $\mathrm{NO_{3u}}$ is used two times. But, it may be puzzling to readers. We have revised this sentence in the manuscript (lines 390-391).

39. Page 16, lines 4-16: Similar to my above comment, please explain how you get the vertical correlation length-scales from Fig. 6.

For the vertical correlation, we use the real values calculated from the forecasting differences in the DA system, but not approximate values from an alternative function. The name of “vertical correlation length-scale” is just a conception to explain the difference between the unbalanced variables and full variables. We have added some explanations in the revised manuscript (lines 397-399, 405-406)

40. Page 16, lines 13-16: I only see very small differences regarding the vertical correlations between the full and unbalanced variables. Perhaps you may wish to modify the text.

The differences of vertical correlation are slight, compared with the difference of horizontal. The main reason is that the vertical correlations are generally affected by the atmospheric boundary layer height. Thus, all vertical correlation decreases rapidly for the level above the boundary layer height. We have added this explanation in the revised manuscript (lines 410-413).

41. Page 17, line 23: Please clarify that DA-balance assimilates the same observations as “DA-full”.

For the DA-full experiments and DA-balance experiments, we use the same observation for the data assimilation. This sentence has been revised (lines 469-470).

42. Page 17, line 25: “WRF” not “WRf”.

Thanks, the sentence has been revised.

43. Page 17, line 28: Please clarify what you mean by “the initial time”.

The initial time means the start time of the model running, which is listed in Table 2.

44. Page 18, lines 4-26: I feel like this discussion slightly misses the main points. In

my opinion, the main point is that the balance constraints can allow observations of a specific species to impact other variables. Even with PM2.5 observations, because the model-simulated PM2.5 is a function of all the control variables, the individual species' fields are adjusted through the BECs, even without a direct observation of the individual species. Thus, without multivariate correlations, an aircraft observation of OC can only impact OC (because the forward operator for OC is only a function of OC), but with the multivariate BECs, an OC observation can now impact OTR or EC. Perhaps you might wish to clarify some aspects of the text along these lines.

Yes. For the BECs without balance constraints, the observation of OC can only impact OC. The crossing effects among species from the BECs with balance constraints. This section has been revised (lines 484-489).

45. Page 19, lines 1-5: I don't believe it is appropriate to describe the smaller RMSEs as "improvements". You're simply looking at fits to observations, which, on their own, do not tell you anything about the relative goodness of your DA system.

The comparison between the analysis PM2.5 against the assimilated observations is known as "sanity check". It can demonstrate the capability of the DA system. In the revised manuscript, we use more data from all nine cases to demonstrate the effects of the DA system. This section has been revised (lines 506-517).

46. Page 19, line 17: The description here of Fig. 11 is incorrect.

Corrected.

47. Page 19, line 19: It should be Fig. 11a, not 1a.

Corrected.

48. Page 19, line 20: Omit "significantly". You can maybe replace it with "substantially".

Thanks, the sentence has been revised.

49. Page 20, lines 16-17: Please clarify what you mean by these lines.

This sentence means the horizontal correlation scales of unbalanced variables are closer than that of full variables. And the vertical correlation scales show similar trend. The sentence has been revised (lines 556-558).

50. Page 20, line 27: Please clarify what you mean by "mutual spread".

The "mutual spread" has been changed to "crossing spread" (line 489).

51. Page 21, line 6: I don't agree with this line. You're only looking at the analysis fits, which does not mean your analysis fields are necessarily better.

This sentence has been removed in the revised manuscript.

52. Page 21, line 20: Please clarify what you mean by a “universal balance constraint”.

The balance constraint in this paper is just a statistical relationship. We hope to find a universal balance that can describe the physical or chemical balanced relationship of aerosol variables, similar with the balance constraint of geostrophic balance or temperature-salinity balance in meteorological or oceanic data assimilation. The sentence has been revised in the manuscript (lines 585-586).

53. Table 1: Suggest also pointing to Eq. (7) in the caption. Also, you should annotate the various species on this figure somehow, because it’s difficult to look back to Eq. (7).

Thanks, we followed this suggestion.

54. Fig. 2 caption: Suggest “NEI05” rather than just “NEI”

Thanks, the sentence has been revised.

55. Fig. 4 caption: In my opinion, this figure isn’t that close to Fig. 3 so I suggest elaborating.

Corrected, thank you .

56. Fig. 5 caption: Suggest pointing to Fig. 4 rather than Fig. 3.

Corrected.

57. Fig. 6: Suggest adding labels of “Height” to the axes.

Corrected.

58. Fig. 7: Suggest adding a unit (meters) to the colorbar.

Corrected.

59. Figs. 8 and 9: The labels above/below the panels are very small. Can these be enlarged?

Corrected.

Background error covariance with balance constraints for aerosol species and applications in variational data assimilation

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Abstract

Balance constraints are important for background error covariance (BEC) in data assimilation to spread information between different variables and produce balance analysis fields. Using statistical regression, we develop a balance constraint for the BEC of aerosol variables and apply it to a three-dimensional variational data assimilation system in the WRF/Chem model. One-month forecasts from the WRF/Chem model are employed for BEC statistics. The cross-correlations between the different species are generally high. The largest correlation occurs between elemental carbon and organic carbon with as large as 0.9. After using the balance constraints, the correlations between the unbalanced variables reduce to less than 0.2. A set of data assimilation and forecasting experiments is performed. In these experiments, surface $PM_{2.5}$ concentrations and speciated concentrations along aircraft flight tracks are assimilated. The analysis increments with the balance constraints show spatial distributions more complex than those without the balance constraints, which is a consequence of the spreading of observation information across variables due to the balance constraints. The forecast skills with the balance constraints show substantial and durable improvements from the 2nd hour to the 16th hour compared with the forecast skills without the balance constraints. The results suggest that the developed balance constraints are important for the aerosol assimilation and forecasting.

Keyword: aerosol species, WRF/Chem, data assimilation, balance constraint, background error covariance

1. Introduction

Aerosol data assimilation in chemical transport models has received an increasing amount of attention in recent years as a basic methodology for improving aerosol analysis and forecasting. In a data assimilation system, the background error covariance (BEC) plays a crucial role in the success of an assimilation process. The BEC and the observation error determine analysis increments from the assimilation process (Derber and Bouttier 1999, Chen et al., 2013).

However, accurate estimation of the BEC remains difficult due to a lack of information about the true atmospheric states and also due to computational requirement arising from the large dimension of the BEC (typically $10^7 \times 10^7$). For a variational data assimilation system, a few methods have been developed to estimate and simplify the expression of the BEC, such as the analysis of innovations, the NMC (National Meteorological Center) and the ensemble-based (Monte Carlo) methods. The NMC method is extensively used in operational atmospheric and meteorology-chemistry data assimilation systems. It assumes that the forecast errors are approximated by differences between pairs of forecasts that are valid at the same time (Parrish and Derber, 1992). Pagowski et al. (2010) estimated the BEC of $PM_{2.5}$ (particles having an aerodynamic diameter less than 2.5 μm) by calculating the differences between the forecasts of 24 and 48 h, and used the estimated BEC in a Grid-point Statistical Interpolation (GSI) system (Wu et al., 2002). Benedetti et al. (2007) estimated the BEC of the sum of the mixing ratios of all aerosol species for an operational analysis and forecast systems at ECMWF (The European Centre for Medium-Range Weather Forecasts). The BEC with multiple species and size bins of aerosols have been calculated and employed in data assimilation. Liu et al. (2011) estimated the BEC with 14 aerosol species in the Goddard Chemistry Aerosol Radiation and Transport scheme of the Weather Research and Forecasting/Chemistry (WRF/Chem) model and applied it to the GSI system. Schwartz et al. (2012) increased the number of the species to 15 based on the study of Liu et al. (2011). Li et al. (2013) estimated the BEC for five species derived from the Model for Simulation Aerosol Interactions and Chemistry (MOSAIC) scheme.

One important role that the BEC plays in meteorological data assimilation is to spread information between different variables to produce balanced analysis fields, which employ balance constraints to convert original variables into new independent variables. Balance constraints have been employed in atmospheric and oceanic data assimilation, such as geostrophic balance or temperature-salinity balance (Bannister, 2008a, 2008b). To incorporate balance constraints, the model variables are usually transformed to balanced and unbalanced parts. The unbalanced parts as control variables are can be assumed independent, and the balanced parts are constrained by balance constraints (Derber and Bouttier, 1999). Instead of using an empirical function as a balance constraint, balance constraints are also derived using regression techniques (Ricci and Weaver, 2005). Although distinct empirical relations between some variables (such as temperature and humidity) may not exist, the regression equation can also be estimated as balance constraints (Chen et al., 2013).

In current aerosol variational data assimilation with multiple variables, balance constraints are not yet incorporated in the BEC. The state variables are assumed to be independent variables without cross-correlation. However, the aerosol species are frequently highly correlated due to their common emission sources and diffusion processes. For example, the correlations in terms of the R-square between elemental carbon and black carbon exceed 0.6 in many locations across Asia and the South Pacific in both urban and suburban locations (Salako et al., 2012), and the correlations between different size bins, such as $PM_{2.5}$ and $PM_{10-2.5}$ (the diameter of particles being between 2.5 and 10 μm), are also generally significant (Sun et al., 2003; Geller et al., 2004). Thus, the cross-correlations between different species or size bins are necessary to produce balanced analysis fields. Cross-correlations spread the observation information from one variable to other variables, which can produce more balanced initial fields. For the data assimilation of the ensemble Kalman filter method, the BEC with balance constraints is assured (Pagowski et al., 2012; Schwartz et al., 2014), although the balance may break down because of localization.

Recently, several studies have suggested that the BEC with balanced cross-correlation should be introduced into aerosol variational data assimilation (Kahnert, 2008; Liu et al., 2011; Li et al., 2013; Saide et al., 2013). Kahnert (2008) exhibited cross-correlations of the seventeen aerosol variables from Multiple-scale Atmospheric Transport and Chemistry (MATCH) Model. He found that the statistical cross-correlations between aerosol components are primarily influenced by the interrelations between emissions and by interrelations due to chemical reactions to a much lesser degree. Saide et al., (2012; 2013) incorporated the capacity to add cross-correlations between aerosol size bins in GSI for assimilating observations of aerosol optical depth (AOD) data. The cross-correlations between the two connecting size bins for each species were considered using recursive filters while, the cross-correlation is not considered for the other size bins that are not connecting.

In this paper, we explore incorporating cross-correlations between different species in BEC using balance constraints. The balance constraints are established using statistical regression. We apply the BEC with the balance constraints to a data assimilation and forecasting system with the MOSAIC scheme in WRF/Chem. The MOSAIC scheme includes a large number of variables with eight species, and flexibility of eight or four size bins. The scheme of four size bins is used in our studies. The four bins are located between 0.039–0.1 μm, 0.1–1.0 μm, 1.0–2.5 μm, and 2.5–10 μm, and the total mass of the first three bins are PM2.5. A 3DVAR system for the MOSAIC (4-bin) scheme has been developed by Li et al. (2013). For comparisons, we employ this 3DVAR system with the same model configurations as employed by Li et al. (2013). The data assimilation and forecasting experiments are performed with a focus on assessing the impact of cross-correlations of the BEC on analyses and forecasts.

The paper is organized as follows: Section 2 describes the 3DVAR system and the formulation of the BEC. Section 3 describes the WRF/Chem configuration and estimates the correlations among the emissions. The statistical characteristics of the BEC, including the regression coefficient of the cross-correlation, are discussed in Section 4. Using the BEC, experiments of assimilating surface $PM_{2.5}$ observations and aircraft observations are discussed in Section 5. Shortcomings, conclusions and future perspectives are presented in Section 6.

2. Data assimilation system and BEC

In this section, we present a formulation of the BEC with cross-correlation between different species using a regression technique. Then, the cost function with the new BEC is derived and the calculating factorization of the BEC is described.

The control variables of the data assimilation are obtained from the MOSAIC (4-bin) aerosol scheme in the WRF/Chem model (Zaveri et al., 2008). The MOSAIC scheme includes eight aerosol species, that is, elemental carbon or black carbon (EC/BC), organic carbon (OC), nitrate (NO_3), sulfate (SO_4), chloride (Cl), sodium (Na), ammonium (NH_4), and other inorganic mass

(OIN). Each species is separated into four bins with different sizes: 0.039–0.1 µm, 0.1–1.0 µm, 1.0–2.5 µm and 2.5–10 µm. The scheme involves 32 aerosol variables with eight species and four size bins. These variables cannot be directly introduced as control variables in an assimilation system in consideration of computational efficiency. The number of variables must be decreased prior to assimilation. Li et al. (2013) have lumped these variables into five species as control variables in the 3DVAR system. The five species consist of EC, OC, NO_3, SO_4 and OTR. Here, OTR is the sum of Cl, Na, NH_4 and OIN. Note that the data assimilation system aims to assimilate the observation of $PM_{2.5}$; only the first three of four size bins are utilized to lump as one control variable for each species.

For a 3DVAR system, the cost function (J), which measures the distance of the state vector to the background and observations, can be written as follows:

$$J(\boldsymbol{x}) = \frac{1}{2}(\boldsymbol{x} - \boldsymbol{x}^{\boldsymbol{b}})^T \mathbf{B}^{-1}(\boldsymbol{x} - \boldsymbol{x}^{\boldsymbol{b}}) + \frac{1}{2}(\boldsymbol{y} - \mathbf{H}\boldsymbol{x})^T \mathbf{R}^{-1}(\boldsymbol{y} - \mathbf{H}\boldsymbol{x}). \qquad (1)$$

Here, $\boldsymbol{x}$ is the vector of the state variables, including EC, OC, NO_3, SO_4 and OTR; $\boldsymbol{x}^{\boldsymbol{b}}$ is the background vector of these five species, which are generated by the MOSAIC scheme; $\boldsymbol{y}$ is the observation vector; $\mathbf{H}$ is the observation operator that maps the model space to the observation space and is assumed to be linear here; $\mathbf{R}$ is the observation error covariance associated with $\boldsymbol{y}$; and $\mathbf{B}$ is the background error covariance associated with $\boldsymbol{x}^{\boldsymbol{b}}$. Eq. (1) is usually written in the incremental form

$$J(\boldsymbol{\delta x}) = \frac{1}{2}\boldsymbol{\delta x}^T \mathbf{B}^{-1}\boldsymbol{\delta x} + \frac{1}{2}(\mathbf{H}\boldsymbol{\delta x} - \boldsymbol{d})^T \mathbf{R}^{-1}(\mathbf{H}\boldsymbol{\delta x} - \boldsymbol{d}), \qquad (2)$$

where $\boldsymbol{\delta x}$ ($\boldsymbol{\delta x} = \boldsymbol{x} - \boldsymbol{x}^b$) is the incremental state variable. The observation innovation vector is known as $\boldsymbol{d} = \boldsymbol{y} - \mathbf{H}\boldsymbol{x}^{\boldsymbol{b}}$. The minimization solution is the analysis increment $\boldsymbol{\delta x}$, and the final analysis is $\boldsymbol{x}^{\boldsymbol{a}} = \boldsymbol{x}^{\boldsymbol{b}} + \boldsymbol{\delta x}$. This analysis is statistically optimal as a minimum error variance estimate (e.g., Jazwinski, 1970; Cohn, 1997).

In Eq. (1) or Eq. (2), $\boldsymbol{x}^{\boldsymbol{b}}$ is a $(N \times m)$ – vector, where N is the number of model grid points, and m is the number of state variables. $\mathbf{B}$ is a symmetric matrix with a dimension of $(N \times m)^2$. For a high-resolution model, the number of vector $\boldsymbol{x}^{\boldsymbol{b}}$ is on the order of 10^7. Therefore, the number of elements in $\mathbf{B}$ is approximately 10^{14}. With this dimension, $\mathbf{B}$ cannot be explicitly manipulated. To pursue simplifications of $\mathbf{B}$, we employ the following factorization

$$\mathbf{B} = \mathbf{DCD}^{\mathbf{T}}, \tag{3}$$

where $\mathbf{D}$ and $\mathbf{C}$ are the standard deviation matrix and the correlation matrix, respectively. $\mathbf{D}$ and $\mathbf{C}$ can be described and separately prescribed after the factorization. $\mathbf{D}$ is a diagonal matrix whose elements include the standard deviation of all state variables in the three-dimensional grids. To reduce the computational cost, we use the average value of standard deviations that are at the same level. Thus, the standard deviation is simplified with vertical levels. $\mathbf{C}$ is a symmetric matrix, having the form

$$\mathbf{C} = \begin{bmatrix} \mathbf{C}_{\mathbf{EC}} & \mathbf{C}_{\mathbf{EC}}^{\mathbf{OC}} & \mathbf{C}_{\mathbf{EC}}^{\mathbf{NO_3}} & \mathbf{C}_{\mathbf{EC}}^{\mathbf{SO_4}} & \mathbf{C}_{\mathbf{EC}}^{\mathbf{OTR}} \\ \mathbf{C}_{\mathbf{OC}}^{\mathbf{EC}} & \mathbf{C}_{\mathbf{OC}} & \mathbf{C}_{\mathbf{OC}}^{\mathbf{NO_3}} & \mathbf{C}_{\mathbf{OC}}^{\mathbf{SO_4}} & \mathbf{C}_{\mathbf{OC}}^{\mathbf{OTR}} \\ \mathbf{C}_{\mathbf{NO_3}}^{\mathbf{EC}} & \mathbf{C}_{\mathbf{NO_3}}^{\mathbf{OC}} & \mathbf{C}_{\mathbf{NO_3}} & \mathbf{C}_{\mathbf{NO_3}}^{\mathbf{SO_4}} & \mathbf{C}_{\mathbf{NO_3}}^{\mathbf{OTR}} \\ \mathbf{C}_{\mathbf{SO_4}}^{\mathbf{EC}} & \mathbf{C}_{\mathbf{SO_4}}^{\mathbf{OC}} & \mathbf{C}_{\mathbf{SO_4}}^{\mathbf{NO_3}} & \mathbf{C}_{\mathbf{SO_4}} & \mathbf{C}_{\mathbf{SO_4}}^{\mathbf{OTR}} \\ \mathbf{C}_{\mathbf{OTR}}^{\mathbf{EC}} & \mathbf{C}_{\mathbf{OTR}}^{\mathbf{OC}} & \mathbf{C}_{\mathbf{OTR}}^{\mathbf{NO_3}} & \mathbf{C}_{\mathbf{OTR}}^{\mathbf{SO_4}} & \mathbf{C}_{\mathbf{OTR}} \end{bmatrix}, \tag{4}$$

where $\mathbf{C}_{\mathbf{EC}}$, $\mathbf{C}_{\mathbf{OC}}$, $\mathbf{C}_{\mathbf{NO_3}}$, $\mathbf{C}_{\mathbf{SO_4}}$ and $\mathbf{C}_{\mathbf{OTR}}$ at diagonal locations are the background error auto-correlation matrices that are associated with each species. They represent the correlation among pairs of grid points for one species. Other submatrices represent the correlations between different species, known as cross-correlations. For example, $\mathbf{C}_{\mathbf{OC}}^{\mathbf{EC}}$ represents the cross-correlations between EC and OC, and $\mathbf{C}_{\mathbf{OC}}^{\mathbf{EC}} = (\mathbf{C}_{\mathbf{EC}}^{\mathbf{OC}})^{\mathbf{T}}$. In Li et al. (2013), these cross-correlations were disregarded, that is, the five species are considered independently and $\mathbf{C}$ is thus a block diagonal matrix.

In this study, the cross-correlations between different species are considered by introducing control variable transforms (Derber and Bouttier, 1999; Barker, 2004; Huang, 2009). We divide the model aerosol variables into balanced components ($\boldsymbol{\delta x_b}$) and unbalanced components ($\boldsymbol{\delta x_u}$):

$$\boldsymbol{\delta x} = \boldsymbol{\delta x_b} + \boldsymbol{\delta x_u}. \tag{5}$$

Note the EC does not need to be divided. There is not unbalanced component for EC that is similar to the variable of vorticity in the data assimilation of ECMWF (Derber and Bouttier, 1999), or the variable of stream function in the data assimilation of MM5 (Barker, 2004). The transformation from unbalanced variables ($\boldsymbol{\delta x_u}$) to full variables ($\boldsymbol{\delta x}$) by the balance operator $\mathbf{K}$ is given by

$$\boldsymbol{\delta x} = \mathbf{K}\boldsymbol{\delta x_u}\,. \tag{6}$$

Eq. (6) can be written as

$$\begin{bmatrix} \boldsymbol{\delta EC} \\ \boldsymbol{\delta OC} \\ \boldsymbol{\delta NO_3} \\ \boldsymbol{\delta SO_4} \\ \boldsymbol{\delta OTR} \end{bmatrix} = \begin{bmatrix} \mathbf{I} & \mathbf{0} & \mathbf{0} & \mathbf{0} & \mathbf{0} \\ \boldsymbol{\rho}_{21} & \mathbf{I} & \mathbf{0} & \mathbf{0} & \mathbf{0} \\ \boldsymbol{\rho}_{31} & \boldsymbol{\rho}_{32} & \mathbf{I} & \mathbf{0} & \mathbf{0} \\ \boldsymbol{\rho}_{41} & \boldsymbol{\rho}_{42} & \boldsymbol{\rho}_{43} & \mathbf{I} & \mathbf{0} \\ \boldsymbol{\rho}_{51} & \boldsymbol{\rho}_{52} & \boldsymbol{\rho}_{53} & \boldsymbol{\rho}_{54} & \mathbf{I} \end{bmatrix} \begin{bmatrix} \boldsymbol{\delta EC} \\ \boldsymbol{\delta OC_u} \\ \boldsymbol{\delta NO_{3u}} \\ \boldsymbol{\delta SO_{4u}} \\ \boldsymbol{\delta OTR_u} \end{bmatrix}, \qquad (7)$$

where $\boldsymbol{\rho}_{\mathrm{ij}}$ is the submatrix of **K**, which represents the statistical regression coefficients between the variables i and j (Chen et al., 2013). Note that $\boldsymbol{\rho}_{\mathbf{ij}}$is a diagonal matrix with the dimension of model grid points. Each model grid point has a regression coefficient. For convenience, we assumed that the elements of $\boldsymbol{\rho}_{\mathrm{ij}}$ is a constant value for all grid points, which are denoted as ρ_{ij} and are calculated by linear regression with all grid points. For example, ρ_{21} can be obtained from the regression equation of OC and EC as

$$\boldsymbol{\delta OC} = \rho_{21}\boldsymbol{\delta EC} + \boldsymbol{\varepsilon}, \qquad (8)$$

where $\boldsymbol{\varepsilon}$ is the residual. $\boldsymbol{\delta EC}$ and $\boldsymbol{\delta OC}$ can be estimated from the forecast differences of 24 h forecasts and 48 h forecasts, similar to the statistics of the BEC. Eq. (8) contains the slope but no intercept. The intercept is nearly zero because $\boldsymbol{\delta EC}$ and $\boldsymbol{\delta OC}$ represent forecast differences that can be considered to be zero mean values. After obtaining ρ_{21}, the balanced part (e.g., the value of the regression prediction) of $\boldsymbol{\delta OC}$ can be obtained by

$$\boldsymbol{\delta OC_b} = \widehat{\boldsymbol{\delta OC}} = \rho_{21}\boldsymbol{\delta EC}\,. \qquad (9)$$

Where $\widehat{\boldsymbol{\delta OC}}$ represents the predicted value of Eq. (8), which is equal to the balanced part ($\boldsymbol{\delta OC_b}$). Remove the $\boldsymbol{\delta OC_b}$ from the full variables to obtain the unbalanced part ($\boldsymbol{\delta OC_u}$), that is, $\boldsymbol{\varepsilon}$ in Eq. (8). Thus, the calculation of $\boldsymbol{\delta OC_u}$ can be written as

$$\boldsymbol{\delta OC_u} = \boldsymbol{\delta OC} - \rho_{21}\boldsymbol{\delta EC}. \qquad (10)$$

Here, $\boldsymbol{\delta OC_u}$ and $\boldsymbol{\delta EC}$ are employed as predictors in the next regression equation to obtain $\boldsymbol{\delta NO_{3b}}$. Then, we can obtain the unbalanced parts of the remaining variables, which are defined as follows:

$$\boldsymbol{\delta NO_{3u}} = \boldsymbol{\delta NO_3} - (\rho_{31}\boldsymbol{\delta EC} + \rho_{32}\boldsymbol{\delta OC_u}), \qquad (11)$$

$$\boldsymbol{\delta SO_{4u}} = \boldsymbol{\delta SO}_4 - (\rho_{41}\boldsymbol{\delta EC} + \rho_{42}\boldsymbol{\delta OC_u} + \rho_{43}\boldsymbol{\delta NO_{3u}}), \qquad (12)$$

$$\boldsymbol{\delta OTR_u} = \boldsymbol{\delta OTR} - (\rho_{51}\boldsymbol{\delta EC} + \rho_{52}\boldsymbol{\delta OC_u} + \rho_{53}\boldsymbol{\delta NO_{3u}} + \rho_{54}\boldsymbol{\delta SO_{4u}}), \qquad (13)$$

The coefficient of determination (R^2) can be employed to measure the fit of these regressions. It can be expressed as

$$R^2 = \frac{\mathrm{SSR}}{\mathrm{SST}}, \tag{14}$$

where SSR and SST are the regression sum of squares and the sum of squares for total, respectively.

These unbalanced parts can be considered to be independent because they are residual and random. $\mathbf{B_u}$ denotes the unbalanced variables of the BEC and can be factorized as

$$\mathbf{B_u} = \mathbf{D_u C_u D_u^T}, \tag{15}$$

where $\mathbf{D_u}$ and $\mathbf{C_u}$ are the standard deviation matrix and the correlation matrix, respectively. $\mathbf{C_u}$ should be a block diagonal without cross-correlations as follows:

$$\mathbf{C_u} = \begin{bmatrix} \mathbf{C_{EC}} & & & & \\ & \mathbf{C_{OCu}} & & & \\ & & \mathbf{C_{NO_{3u}}} & & \\ & & & \mathbf{C_{SO_{4u}}} & \\ & & & & \mathbf{C_{OTRu}} \end{bmatrix}. \tag{16}$$

According the definition of the BEC,

$$\mathbf{B} = \langle(\boldsymbol{\delta x})(\boldsymbol{\delta x}^{\mathrm{T}})\rangle. \tag{17}$$

And $\mathbf{B_u}$ can be written as

$$\mathbf{B_u} = \langle(\boldsymbol{\delta x}_{\mathbf{u}})(\boldsymbol{\delta x}_{\mathbf{u}}{}^{\mathbf{T}})\rangle. \tag{18}$$

Using Eq. (6), Eq. (17) and Eq. (18), the relationship between $\mathbf{B}$ and $\mathbf{B_u}$ is

$$\mathbf{B} = \mathbf{K B_u K^T}. \tag{19}$$

$\mathbf{B}^{\frac{1}{2}}$ and $\mathbf{B}_{\mathbf{u}}^{\frac{1}{2}}$ are defined as the square root of $\mathbf{B}$ and the square root of $\mathbf{B_u}$, respectively. Their transformation is

$$\mathbf{B}^{\frac{1}{2}} = \mathbf{K B}_{\mathbf{u}}^{\frac{1}{2}}. \tag{20}$$

Using Eq. (15), Eq. (20) can be written as follows:

$$\mathbf{B}^{\frac{1}{2}} = \mathbf{K D_u C}_{\mathbf{u}}^{\frac{1}{2}}. \tag{21}$$

Generally, a transformed cost function of Eq. (2) is expressed as a function of a preconditioned state variable:

$$J(\boldsymbol{\delta z}) = \frac{1}{2}\boldsymbol{\delta z}^T\boldsymbol{\delta z} + \frac{1}{2}\left(\mathbf{HB}^{\frac{1}{2}}\,\boldsymbol{\delta z} - \boldsymbol{d}\right)^T \mathbf{R}^{-1}\left(\mathbf{HB}^{\frac{1}{2}}\,\boldsymbol{\delta z} - \boldsymbol{d}\right). \tag{22}$$

Here, $\boldsymbol{\delta z} = \mathbf{B}^{-\frac{1}{2}}\,\boldsymbol{\delta x}$. Using Eq. (21), Eq. (22) can be written as

$$J(\boldsymbol{\delta z}) = \frac{1}{2}\boldsymbol{\delta z}^T\boldsymbol{\delta z} + \frac{1}{2}\left(\mathbf{HKD_u C_u^{\frac{1}{2}}}\,\boldsymbol{\delta z} - \boldsymbol{d}\right)^T \mathbf{R}^{-1}\left(\mathbf{HKD_u C_u^{\frac{1}{2}}}\,\boldsymbol{\delta z} - \boldsymbol{d}\right). \tag{23}$$

Eq. (23) is the last form of the cost function with the cross-correlation of $\mathbf{B}$.

According to Li et al. (2013), the correlation matrix of the unbalanced parts ($\mathbf{C_u}$) is factorized as

$$\mathbf{C_u} = \mathbf{C_{ux}} \otimes \mathbf{C_{uy}} \otimes \mathbf{C_{uz}}. \tag{24}$$

Here, $\otimes$ denotes the Kronecker product, and $\mathbf{C_{ux}}$, $\mathbf{C_{uy}}$ and $\mathbf{C_{uz}}$ represent the correlation matrices between gridpoints in the x direction, the y direction, and the z direction, respectively, with the sizes $n_x \times n_x$, $n_y \times n_y$, and $n_z \times n_z$, respectively. Here, n_x, n_y and n_z represent the numbers of grid points in the x direction, y direction, and z direction, respectively. This factorization can decrease the size of the dimension of $\mathbf{C_u}$. Another desirable property of Eq. (24) is

$$\mathbf{C_u^{\frac{1}{2}}} = \mathbf{C_{ux}^{\frac{1}{2}}} \otimes \mathbf{C_{uy}^{\frac{1}{2}}} \otimes \mathbf{C_{uz}^{\frac{1}{2}}} \tag{25}$$

$\mathbf{C_{ux}}$ and $\mathbf{C_{uy}}$ are expressed by Gaussian functions, and $\mathbf{C_{uz}}$ is directly computed from the proxy data. They will be discussed in Sec 4.2.

3. WRF/Chem configuration and cross-correlations of emission species

In this section, we describe the configuration of WRF/Chem, whose forecasting products will be employed in the following BEC statistics and data assimilation experiments. In addition, the cross-correlations of emission species from the WRF/Chem emission data are investigated to understand the cross-correlation between different species of the BEC.

3.1 WRF/Chem configuration

WRF/Chem (V3.5.1) is employed in our study. This is a fully coupled online model with a regional meteorological model that is coupled to aerosol and chemistry models (Grell et al., 2005). The model domain with three spatial domains is shown in Figure 1. The horizontal grid spacing for these three domains are 36 km (80×60 points), 12 km (97×97 points), and 4 km (144×96 points), respectively. The outer domain spans southern California and the innermost domain encompasses Los Angeles. All domains have 31 vertical levels with the top at 50 hPa. The vertical grid is stretched to place the highest resolution in the lower troposphere. The discussion of the BEC and the emissions presented in this paper will be confined to the innermost domain. The initial meteorology conditions for WRF/Chem are prepared using the North American Regional Reanalysis (NARR) (Mesinger et al. 2006). The meteorology boundary conditions and sea surface temperatures are updated at each initialization. For the forecast running, the initial meteorological conditions are obtained from the NARR data. The initial aerosol conditions are obtained from the former forecast. The emissions are derived from the National Emission Inventory 2005 (NEI'05) for both aerosols and trace gases (Guenther et al., 2006). For more details, the readers are referred to Li et al. (2013).

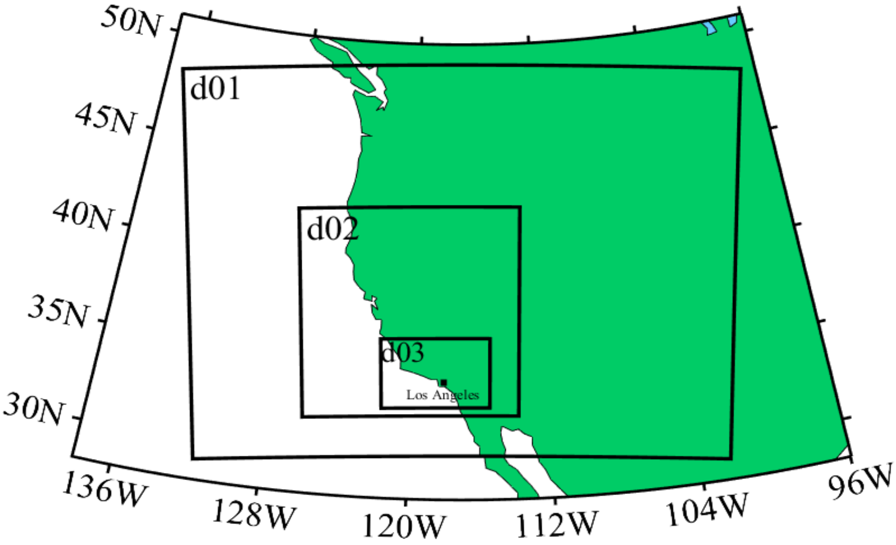


Figure 1. Geographical display of the three-nested model domains. The innermost domain covers the Los Angeles basin; the black point denotes the location of Los Angeles.

3.2 Cross-correlations of emission species

The emission source is necessary for running the WRF/Chem model. It is an important factor for the distribution of the aerosol forecasts. The analysis of the correlations among the emission species can help us to understand the BEC statistics. The emission species is derived from the emission file that is produced by the NEI'05 data for each model domain. Only the emission data for the innermost domain is used to calculate the correlation among the emission species. The emission file contains 37 variables, including gas species and aerosol species. An aerosol species also comprises a nuclei mode and accumulation model species (Peckam et al., 2013). From these aerosol emission species, five lumped aerosol species are calculated, which is consistent with the variables in the data assimilation. These five lumped species are E_EC (sum of the nuclei mode and the accumulation mode of elemental carbon $PM_{2.5}$), E_ORG (sum of the nuclei mode and the accumulation mode of organic $PM_{2.5}$), E_NO3 (sum of the nuclei mode and the accumulation mode of nitrate $PM_{2.5}$), E_SO4 (sum of the nuclei mode and the accumulation mode of sulfate $PM_{2.5}$), and E_PM25 (sum of the nuclei mode and the accumulation mode of unspeciated primary $PM_{2.5}$).

Figure 2 shows the cross-correlations of the five lumped aerosol emission species. All cross-correlations exceed 0.5. This result reveals that the emission species are correlated, which may be attributed to the common emission sources and diffusion processes that are controlled by the same atmospheric circulation. The most significant cross-correlation is between E_EC and E_ORG with a value of approximately 0.8. This high correlation demonstrates that the emission distributions of these two species are very similar. Their emissions are primary in urban and suburban areas with small emissions in rural areas and along roadways (not shown). As shown in Fig. 2, the lowest cross-correlation is between E_ORG and E_SO4; the latter emissions are primary in the urban and suburban areas with few emissions in rural areas and roadways (not shown).

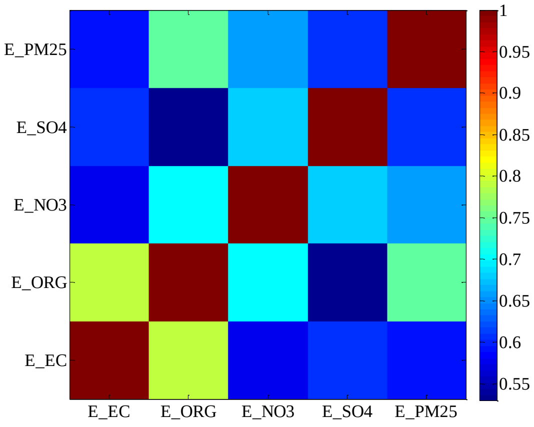


Figure 2. Cross-correlations between emission species of E_EC, E_ORG, E_NO3, E_SO4 and E_PM25. The emission species data are derived from the NEI’05 emissions set for the innermost domain of the WRF/Chem model

4 Balance constraints and BEC statistics

With the configuration of the WRF/Chem model described in Section 3.1, forecasts for one month (from 00UTC of May 15 to 00UTC of June 14, 2010) were performed for the balance constraints and the BEC statistics. Forecast differences between 24 h forecasts and 48 h forecasts are available at 00UTC. Thirty forecast differences are employed as inputs in the NMC method. For this method, 30 forecast differences are sufficient; however, a longer time series may be more beneficial for the BEC statistics (Parrish and Derber, 1992).

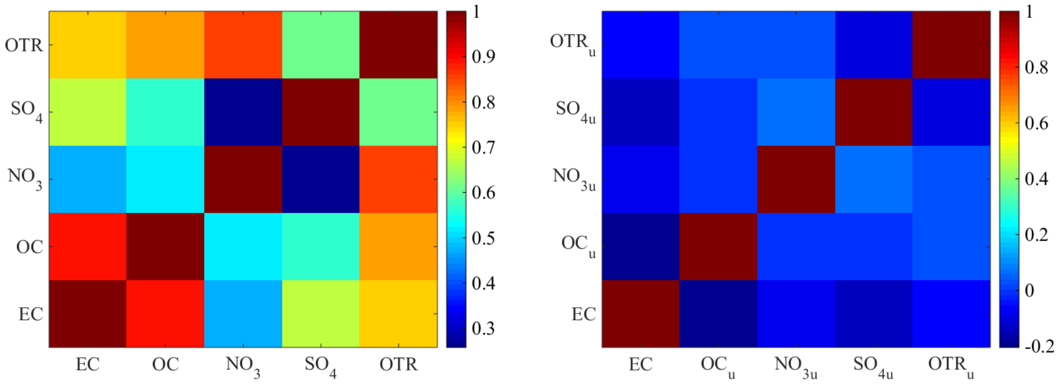
4.1 Balance regression statistics

Using the 30 forecast differences between 24 h and 48 h forecasts, we can obtain $\boldsymbol{\delta EC}$, $\boldsymbol{\delta OC}$, $\boldsymbol{\delta NO_3}$ $\boldsymbol{\delta SO_4}$ and $\boldsymbol{\delta OTR}$. The size of these variables is $(N \times 30)$, where N is the number of model grid points. We put these data into Eqs. (6-13) to calculate the regression coefficients of ρ_{ij} and the unbalanced parts of the variables. Note the process of calculation should be step by step, since the latter equation will use the unbalanced parts of former equations. Table 1 shows the regression coefficients whose column and row are consistent with $\rho_{i,j}$ in Eq. (7). The last column in Tab. 1 is the coefficient of determination (R^2) of the regression equations. For the regression equation of OC, the regression coefficient is 0.90 and the coefficient of determination of Eq. (7) is 0.86, which indicates that EC and OC are highly correlated and their mass concentration scales are approximate. Their correlation is similar to the correlation of the stream function and velocity potential; thus, we set them as the first and second variables in the regression statistics. For the regression equation of $\mathrm{NO_3}$, the regression coefficients of EC and $\mathrm{OC_u}$ are 4.01 and 3.76, respectively, because the mass concentration scale of $\mathrm{NO_3}$ exceeds the mass concentration scales of EC and $\mathrm{OC_u}$. The coefficient of determination is only 0.32, which indicates that the correlations between $\mathrm{NO_3}$ and EC and between $\mathrm{NO_3}$ and $\mathrm{OC_u}$ are weak. This result reveals that the forecast errors of $\mathrm{NO_3}$ differ from the forecast errors of EC and $\mathrm{OC_u}$. A possible reason is that $\mathrm{NO_3}$ is the secondary particle that is primarily derived from the transformation of $\mathrm{NO_x}$, but EC and $\mathrm{OC_u}$ are derived from direct emissions. Similar to $\mathrm{NO_3}$, $\mathrm{SO_4}$ is also primarily derived from the transformation of $\mathrm{SO_2}$ and the coefficient of determination for $\mathrm{SO_4}$ is also low. For the last variable OTR, the maximum coefficient of determination is 0.96 because OTR includes some different compositions that are correlated with the first four variables.

Table 1 Regression coefficients of balance operator $\mathbf{K}$ and the coefficient of determination (regression coefficients correspond to ρ_{ij} in Eq. (7))

species	regression coefficient (ρ)					coefficient of determination (R^2)
EC	1					/
OC	0.90	1				0.86
NO_3	4.01	3.76	1			0.32
SO_4	1.35	-0.21	-3.15	1		0.48
OTR	2.93	2.35	0.28	0.60	1	0.96

Figure 3 shows the cross-correlations of the five full variables and the unbalanced variables. In Fig. 3a, the cross-correlations of the full variables exceed 0.3 and most of them exceed 0.5. In Fig. 3b, however, the cross-correlations of the unbalanced variables are less than 0.2. Some of the cross-correlations are close to zero, which indicates that these unbalanced variables are approximatively independent and can be employed as control variables in the data assimilation system.



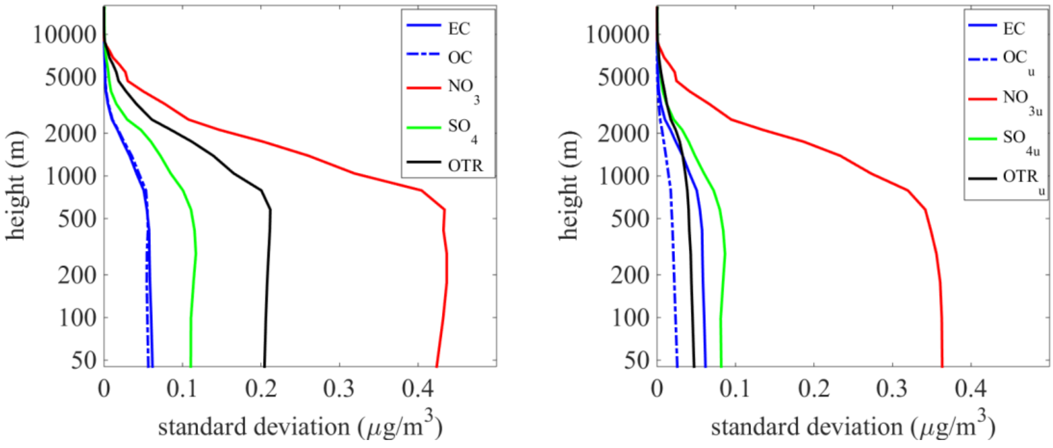
(a) full variables

(b) unbalanced variables

Figure 3. Cross-correlations between the five variables of the BEC. These variables are (a) full variables and (b) unbalanced variables of EC, OC, NO_3, SO_4 and OTR.

4.2 BEC statistics

Using the original full variables and the unbalanced variables obtained by the regression equations, the BEC statistics are obtained. Figure 4 shows the vertical profiles of the standard deviations of the original $\mathbf{D}$ and the unbalanced $\mathbf{D_u}$. In Fig. 4a, the original standard deviation of NO_3 is the largest value, whereas the smallest value is OC, whose profile is close to the profile of EC. All profiles show a significant decrease at approximately 800 m because the aerosol particulates are usually limited under the boundary level. In Fig. 4b, all standard deviations decrease in different degree, with the exception of EC, which remains as the control variable in the unbalanced BEC statistics. Note that the standard deviation of OTR_u decreases by approximately 80% compared with NO_{3u}, which decreases by approximately 10%. This result is attributed to the small coefficient of determination for the regression of NO_3 (in Tab. 1), which indicates that a small portion of NO_3 can be predicted by the regression and a large portion is an unbalanced component. In contrast with NO_3, a small portion of OTR is the unbalanced component.

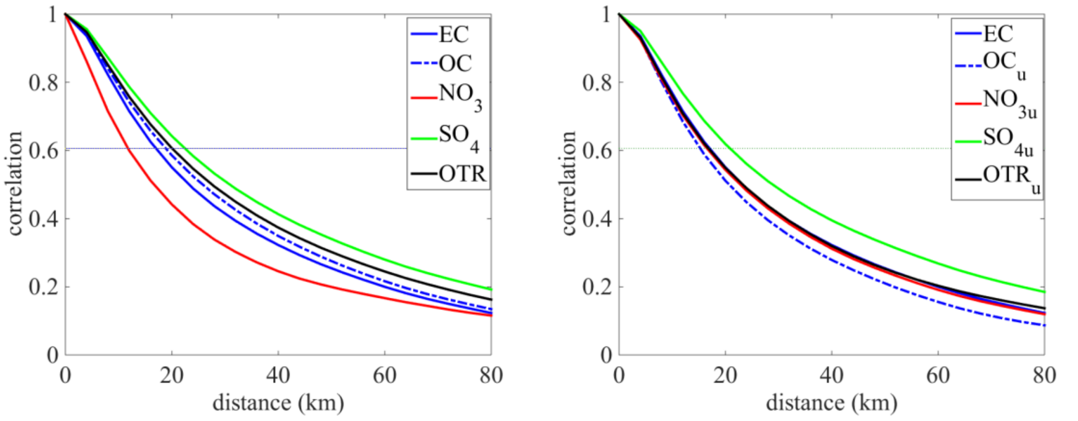


(a) full variables

(b) unbalanced variables

Figure 4. Vertical profiles of the standard deviation of the variables. (a) full variables and (b) unbalanced variables

For the correlation matrix of $\mathbf{C}$ and $\mathbf{C_u}$, they are factorized as three independent one-dimensional correlation matrices in Eq. (24). The horizontal correlation $\mathbf{C_x}$ or $\mathbf{C_y}$ is approximately expressed by a Gaussian function. The correlation between two points r_1 and r_2 can be written as $e^{-\frac{(r_2-r_1)^2}{2L_S^2}}$, where L_S is the horizontal correlation scale and is a constant value for $\mathbf{C_x}$ and $\mathbf{C_y}$, which are considered to be isotropic (Li et al., 2013). This scale can be estimated by the curve of the horizontal correlations with distances. Figure 5 shows the curves of the horizontal correlations for the five control variables. For the full variables (Fig. 5a), the sharpest decrease in the curves is observed for NO_3 and the slowest decrease in the curves is observed for SO_4. We assume that the decline curve is according to the Gaussian function. Then the intersection of the decline curve and the line of $e^{-\frac{1}{2}}\ (\approx 0.61)$ can be approximately as the value of horizontal correlation scale. The horizontal correlation scales of EC, OC, NO_3, SO_4 and OTR are 25 km, 27 km, 20 km, 30 km and 28 km, respectively. For the unbalanced variables (Fig. 5b), their curves are closer than the curves of the full variables. The correlation scales of EC, OC_u, NO_{3u}, SO_{4u} and OTR_u are 25 km, 23 km, 24 km, 28 km and 25 km, respectively. These results suggest that the unbalanced variables are expressed by some common factors such as EC, OC_u and NO_{3u}, in the regression equations of Eqs. (10-13), which produces consistent horizontal correlation scales.

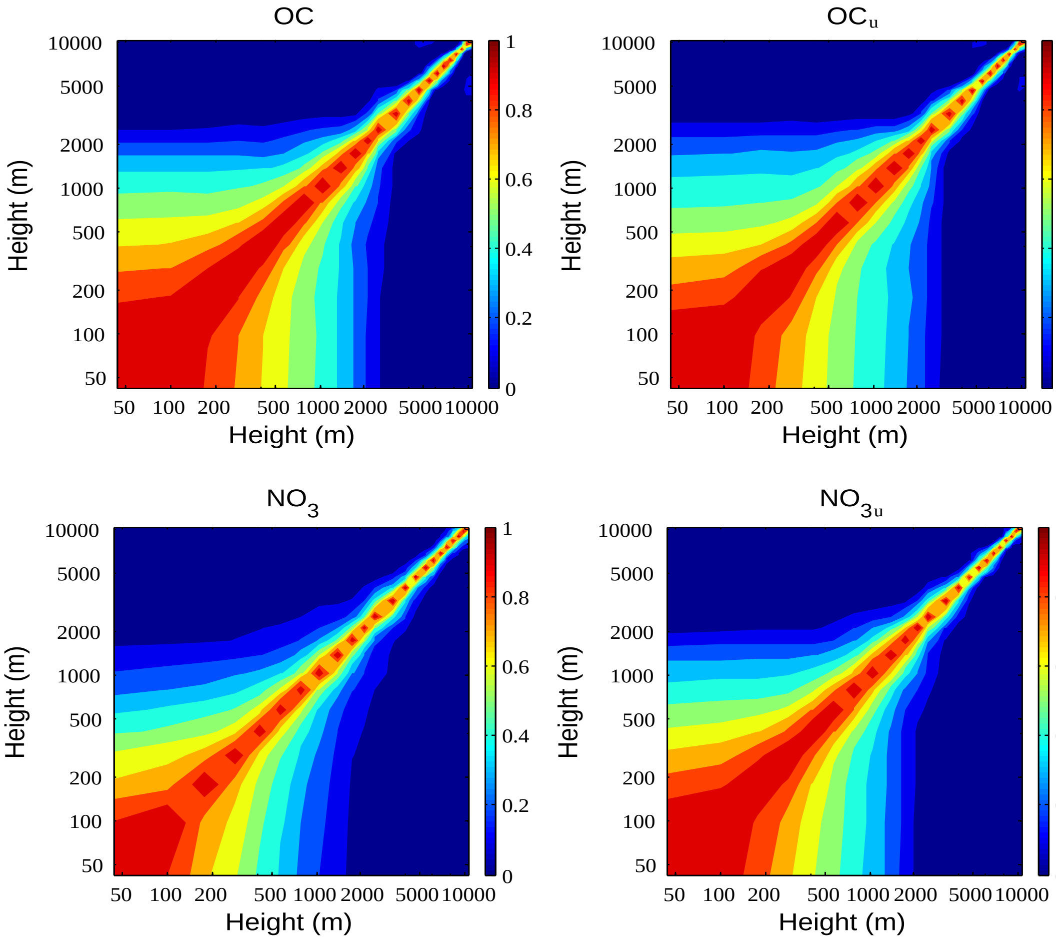


(a) full variables

(b) unbalanced variables

Figure 5. Same as Figure 4, with the exception of the horizontal auto-correlation curves of the variables. The horizontal thin line is the reference line of $e^{-\frac{1}{2}}\ (\approx 0.61)$ for determining the horizontal correlation scales.

For the vertical correlation between $\mathbf{C_z}$ and $\mathbf{C_{uz}}$, they are directly estimated using the forecasting differences in the data assimilation system, but not estimated from a approximately alternative function. Because it is only an $n_z \times n_z$ matrix. Figure 6 shows the vertical correlation matrices $\mathbf{C_z}$ and $\mathbf{C_{uz}}$ for the full variables (left column) and the unbalanced variables (right column), respectively. A common feature of both the full variables and the unbalanced variables is the significant correlation between the levels of the boundary layer height, which is consistent with the profile of the standard deviation in Fig. 4. Some weak adjustments to the correlations between the full and unbalanced variables are made. For example, the correlation of NO_{3u} is stronger than the correlation of NO_3 between the boundary layers. Similar with the analysis of horizontal correlation scale, the vertical correlation scale of NO_{3u} is larger than the vertical correlation scale of NO_3. Conversely, the vertical correlation scale of OTR_u is smaller than the vertical correlation scale of OTR. These results demonstrate that the vertical correlations for the unbalanced variables are more consistent than the vertical correlations of the full variables, which is similar to the adjustments to the horizontal correlation scale. Note that the differences of vertical correlation are slight, compared with the difference of horizontal. The main reason is that the vertical correlations are generally affected by the atmospheric boundary layer height. Thus, all vertical correlation decreases rapidly for the levels above the boundary layer height.



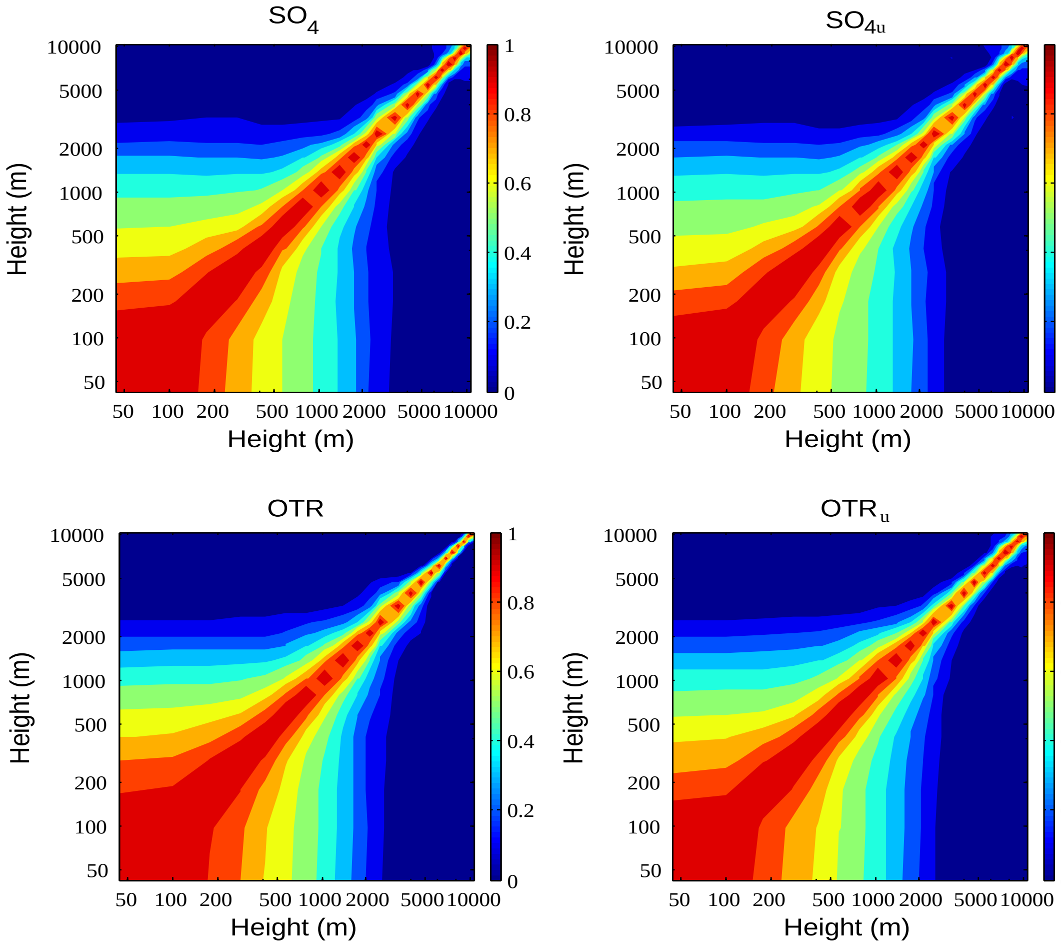


Figure 6. Vertical correlations of the five variables of the BEC. The left column represents the full variables, and the right column represents the unbalanced variables.

5. Application to data assimilation and prediction

To exhibit the effect of the balance constraint of the BEC, the data assimilation experiments and 24-h forecasts for nine cases are run using WRF/Chem model. The surface $PM_{2.5}$ and aircraft-speciated observations are assimilated using different BEC, and the evaluations are presented for the data assimilation and subsequent forecasts. Three basic statistical measures including mean bias (BIAS), root mean square error (RMSE) and correlation coefficient (CORR) are utilized for the evaluations.

5.1 Observation data and experiment scheme

Two types of observation data are employed in our experiments. The first type of observation data consists of hourly surface $PM_{2.5}$ concentrations from the California Air Resources Board (ARB). There are 42 surface PM2.5 monitoring sites existed in the innermost domain of the

WRF/Chem model (Fig. 7). The second type of observation data is the speciated concentration along the aircraft flight track. The aircraft observations were investigated from the California Research at the Nexus of Air Quality and Climate Change (CalNex) field campaign in 2010. Nine flights data around Los Angeles from 15 May to14 June, 2010 are selected as the cases of data assimilation. Table 2 shows the start time and end time of each flight. The species of the aircraft observations include OC, NO_3, SO_4 and NH_4. Note that NH_4 is not a control variable; thus, the aircraft observation of NH_4 is disregarded in the data assimilation. Because the particle size of the aircraft observations is less than 1.0 μm, some adjustments to the flight observations are made according to the ratios between the concentration under 2.5 μm and the concentration under 1.0 μm for each species using model products. With the ratios multiplied by the aircraft observed concentrations, the speciated concentrations under 2.5 μm can be obtained.

Table 2 The periods of flight during CalNex 2010 and the initial time of assimilation

Number of cases	Start time of flight	End time of flight	Initial time of assimilation
1	18:00 UTC, May 16	01:42 UTC, May 17	00:00 UTC, May 17
2	17:28 UTC, May 19	00:10 UTC, May 20	18:00 UTC, May 19
3	17:28 UTC, May 21	00:10 UTC, May 21	18:00 UTC, May 21
4	23:08 UTC, May 24	05:23 UTC, May 25	00:00 UTC, May 25
5	01:59 UTC, May 30	07:45 UTC, May 30	06:00 UTC, May 30
6	05:00 UTC, May 31	10:54 UTC, May 31	06:00 UTC, May 31
7	07:59 UTC, June 2	14:09 UTC, June 2	12:00 UTC, June 2
8	07:59 UTC, June 3	14:041 UTC, June 3	12:00 UTC, June 3
9	17:56 UTC, June 14	23:35 UTC, June 14	18:00 UTC, June 14

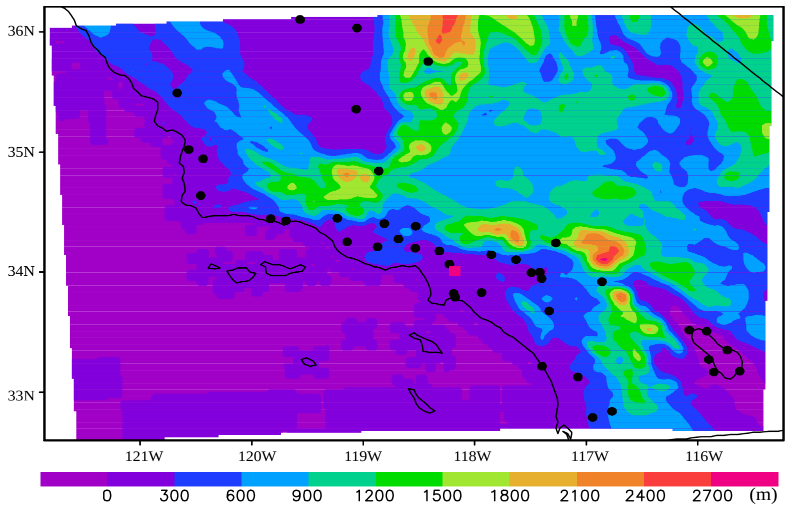
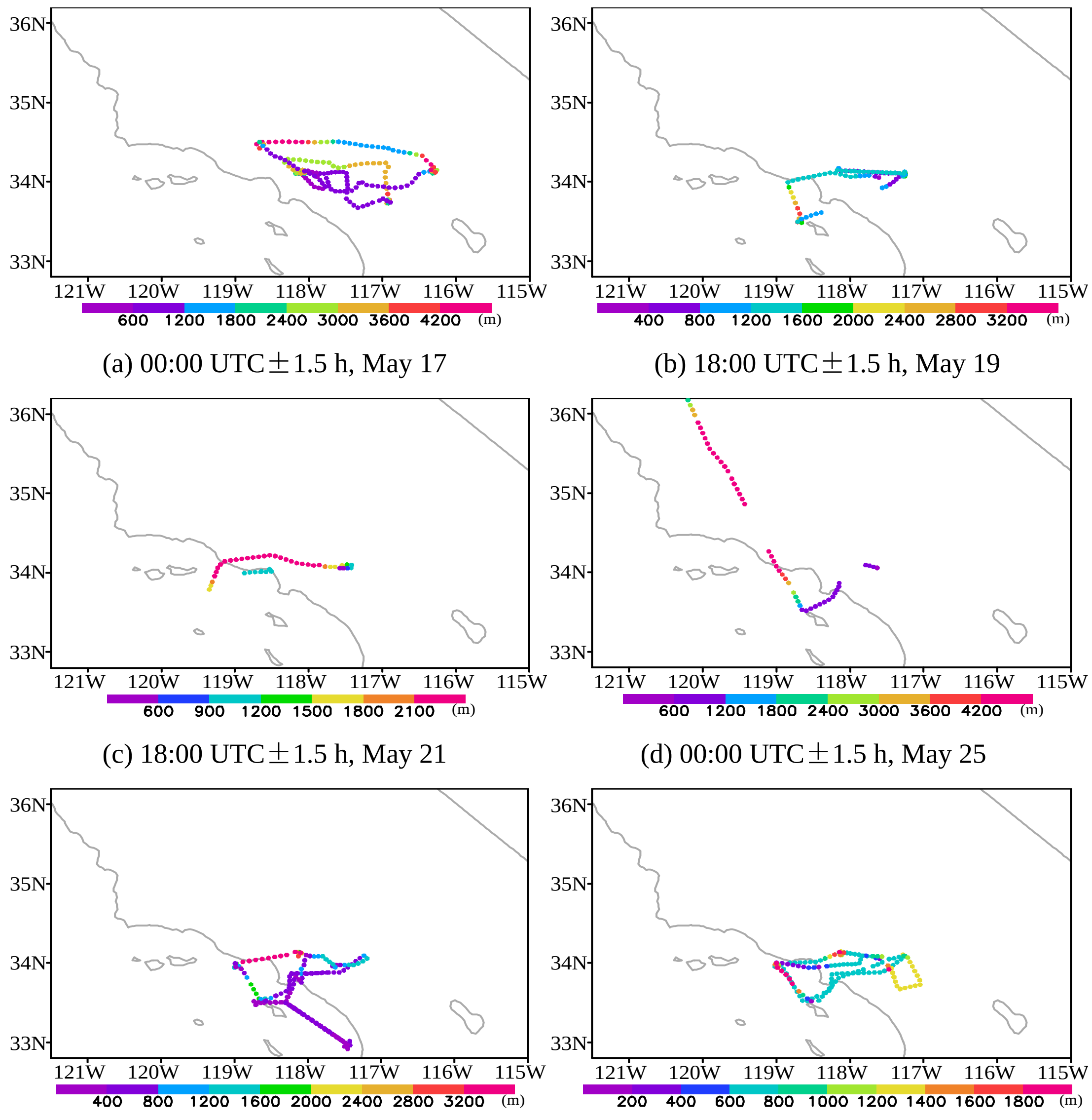


Figure 7. The topography of the innermost domain and the locations of surface monitoring stations (black dots). The red square is the location of Los Angeles


36N
35N
34N
33N
121W
120W
119W
118W
117W
116W
115W
600 1200 1800 2400 3000 3600 4200 (m)
400 800 1200 1600 2000 2400 2800 3200 (m)
600 900 1200 1500 1800 2100 (m)
600 1200 1800 2400 3000 3600 4200 (m)
400 800 1200 1600 2000 2400 2800 3200 (m)
200 400 600 800 1000 1200 1400 1600 1800 (m)

(a) 00:00 UTC ± 1.5 h, May 17

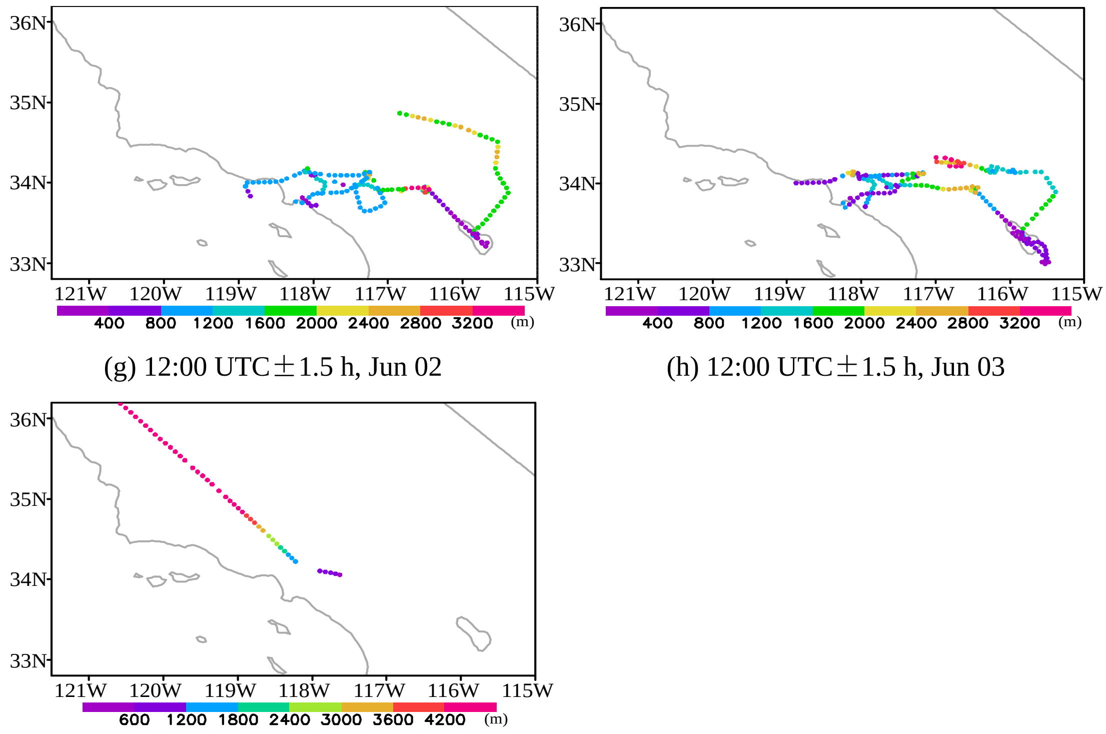
(b) 18:00 UTC ± 1.5 h, May 19

(c) 18:00 UTC ± 1.5 h, May 21

(d) 00:00 UTC ± 1.5 h, May 25

(e) 06:00 UTC ± 1.5 h, May 30

(f) 06:00 UTC ± 1.5 h, May 31


36N
35N
34N
33N
121W 120W 119W 118W 117W 116W 115W
400 800 1200 1600 2000 2400 2800 3200 (m)
600 1200 1800 2400 3000 3600 4200 (m)

(g) 12:00 UTC±1.5 h, Jun 02

(h) 12:00 UTC±1.5 h, Jun 03

(i) 18:00 UTC±1.5 h, Jun 14

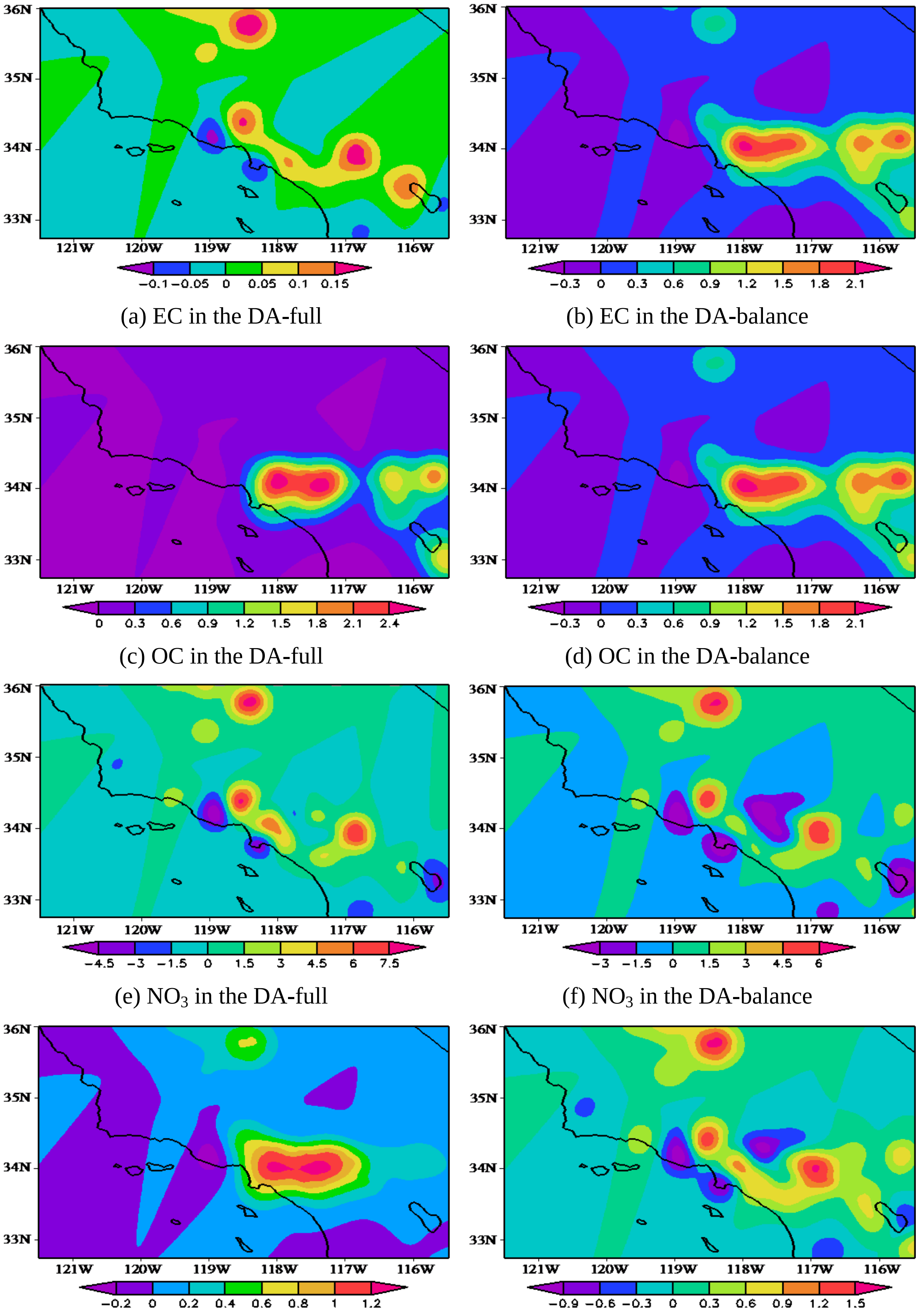
Figure 8. Aircraft flight tracks during the time window of data assimilation for nine cases. The color of the track indicates the aircraft height.

The initial time of data assimilation cases are designed according to the period of flights, showed in Table 2. The time window of assimilation for the flight data is ±1.5h, though some flight times do not completely cover the time windows. Figure 8 shows the aircraft tracks during the time window of data assimilation. It is obvious that the aircarft data on May 21, May 25 and June 14 are relative few as the tracks are almost outside of the study domain. For the surface data, it is only the observations at the initial time are assimilated. For each case, three parallel experiments are performed. The first experiment is the control experiment without aerosol data assimilation, which is frequently known as a free run and denoted as Control. The second experiment is a data assimilation experiment that assimilates surface $PM_{2.5}$ and aircraft observations using the full variables without balance constraints; it is denoted as DA-full. The third experiment is also a data assimilation experiment that also assimilates surface $PM_{2.5}$ and aircraft observations, but employs the unbalanced variables as control variables conducted by the balanced constraint; it is denoted as DA-balance. The backgrounds for DA-full and DA-balance are the forecasting results from the previous runs without DA. These previous forecasting results have been obtained when we run the model for the BEC statistics. The observation error is the half of standard deviation of the original background variable, and a vertical profile of observation errors is applied with the average profile of standard deviation of the background variable. In each experiment, a 24-h forecasting is run using the WRF/Chem model with the same configuration described in Section 3.1, and the case on June 3, 2010 is presented in detail as an example.

5.2 Increments of data assimilation

Figure 9 shows the horizontal increments of EC, OC, NO_3, SO_4 and OTR at the first model level for the DA-full (left column) and DA-balance experiments (right column) of the case on June 3, 2010. In the DA-full experiment, the increment of EC and OTR (Fig. 9a and 9i) are similar. They are obtained from the surface $PM_{2.5}$ observations because no direct aircraft observations correspond to these two variables. In the DA-balance experiment, significant adjustments are made to the increments of EC (Fig. 9b) under the action of the balance constraints. The observations of OC affect greatly the increments of EC for thee high cross-correlation between EC and OC. Thus the increments of EC are similar with the increments of OC. Similarly, significant adjustments are made to the increment of OTR (Fig. 9j), though there are not the species observation of OTR. There are also some slight adjustments for the increments of OC, NO_3 and SO_4 for the crossing spread among species.

Figure 10 shows the vertical increments along 35.0 N for the DA-full and DA-balance experiments. Similar to Fig. 9, the increments of EC and OTR (Fig. 10a and 10i) spread upward from the surface in the DA-full experiment, which are obtained from the surface $PM_{2.5}$ observation. In the DA-balance, the increments of EC and OTR (Fig. 10b and 10j) exhibit observation information from the aircraft height at approximately 500 m, and the value of the increments show significant increases. The distributions of the increments for these five variables in the DA-balance (Fig. 10, right column) generally tend to coincide compared with the distributions of the increments in the DA-full (Fig. 10, left column). The results of the DA-balance are reasonable due to the influence of each other across the balance constraints.



(a) EC in the DA-full

(b) EC in the DA-balance

(c) OC in the DA-full

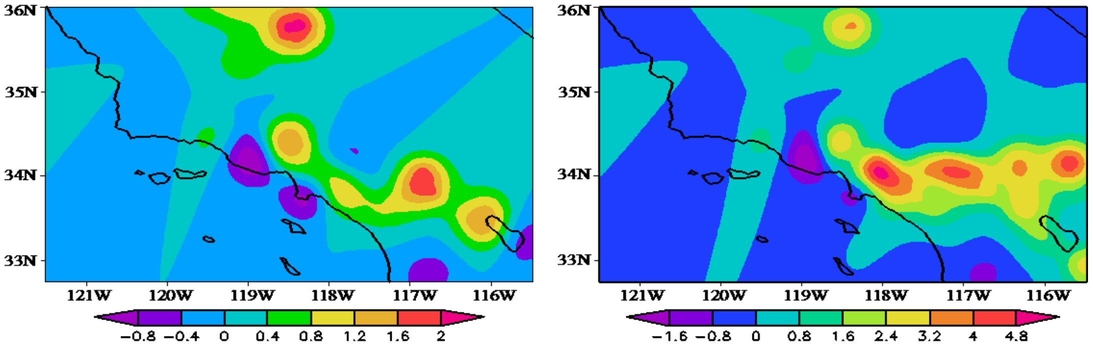
(d) OC in the DA-balance

(e) NO_3 in the DA-full

(f) NO_3 in the DA-balance

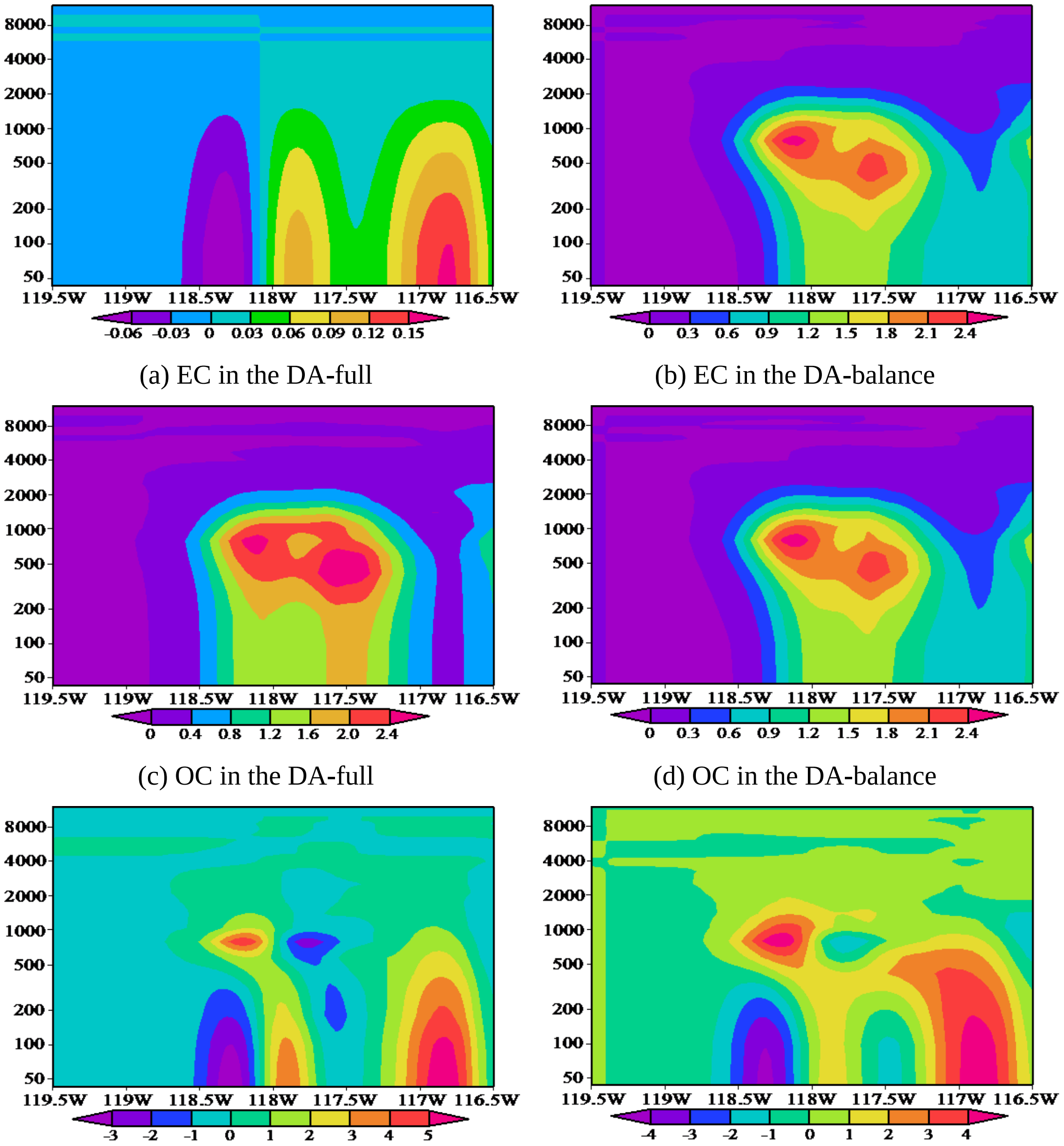
(g) SO_4 in the DA-full

(h) SO_4 in the DA-balance



(i) OTR in the DA-full (j) OTR in the DA-balance

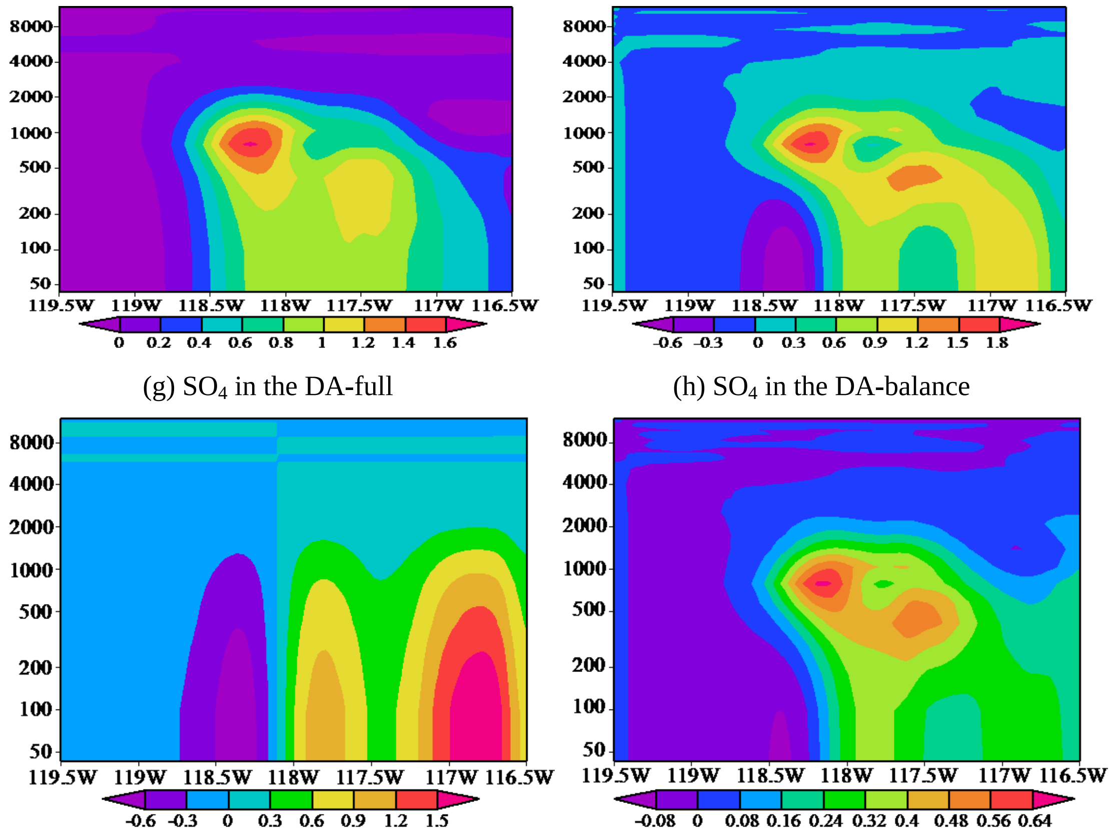
Figure 9. Surface distributions of increments of the five variables of EC, OC, NO_3, SO_4 and OTR at 12:00 UTC on June 3, 2010. The left column and right column are from DA-full and DA-balance, respectively.



(a) EC in the DA-full (b) EC in the DA-balance

(c) OC in the DA-full (d) OC in the DA-balance

(e) NO_3 in the DA-full (f) NO_3 in the DA-balance



(g) SO_4 in the DA-full

(h) SO_4 in the DA-balance

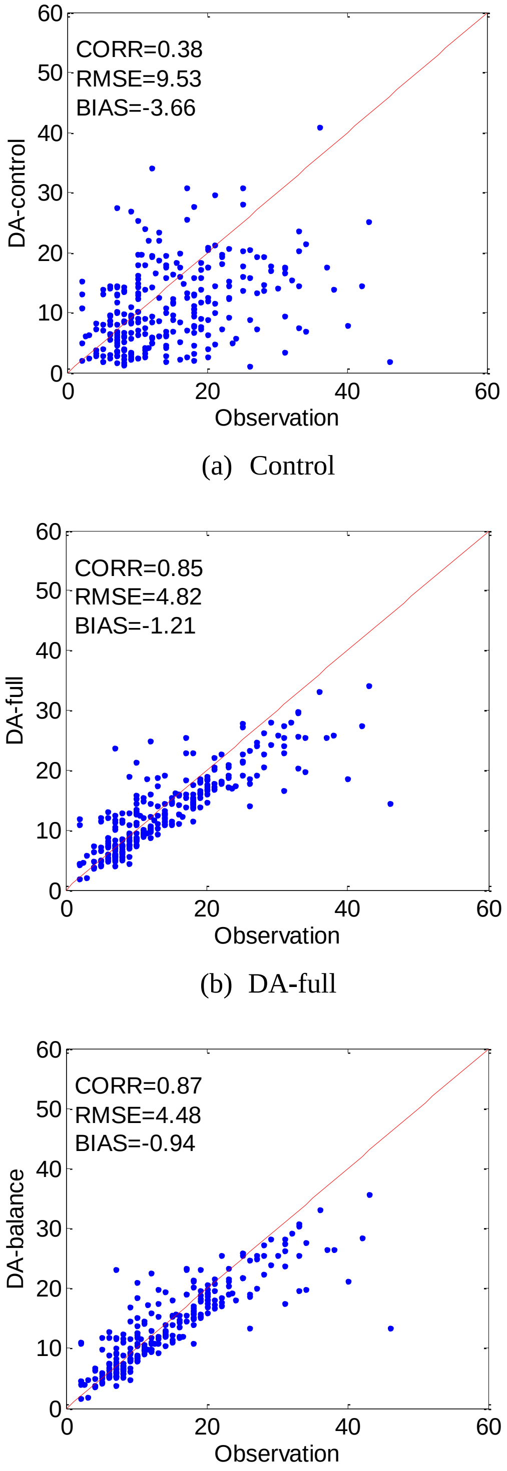
(i) OTR in the DA-full

(j) OTR in the DA-balance

Figure 10. Same as Figure 9, with the exception of the vertical sections along 35 N.

5.3 Evaluation of data assimilation and forecasts

Figure 11 shows the scatter plots of the initial model fields versus the surface observation for all nine cases. In Fig. 11a, the simulated concentrations of the Control experiment display a significant underestimation with a BIAS of -3.66 μg/m^3. The mean concentration of Control is 10.90 μg/m^3, about 25.1% lower than observed mean concentrations (14.56 μg/m^3). In the DA-full and DA-balance experiments, there are remarkable increases for the simulated concentrations, and the BIASs reduce to as small as -1.21 and -0.94 μg/m^3. The RMSE is 9.53 μg/m^3 in the Control experiment. The RMSE reduces to 4.82 and 4.48 μg/m^3 in the DA-full and DA-balance experiment, respectively. There are also significant improvements for the CORR in the DA-full and DA-balance experiments, compared with the Control experiment. Furthermore, these three statistical measures of the DA-balance experiments show some slight improvement, compared with that of the DA-full experiments. The result demonstrates that more observation information spread by balance constraints can improve assimilation performance.



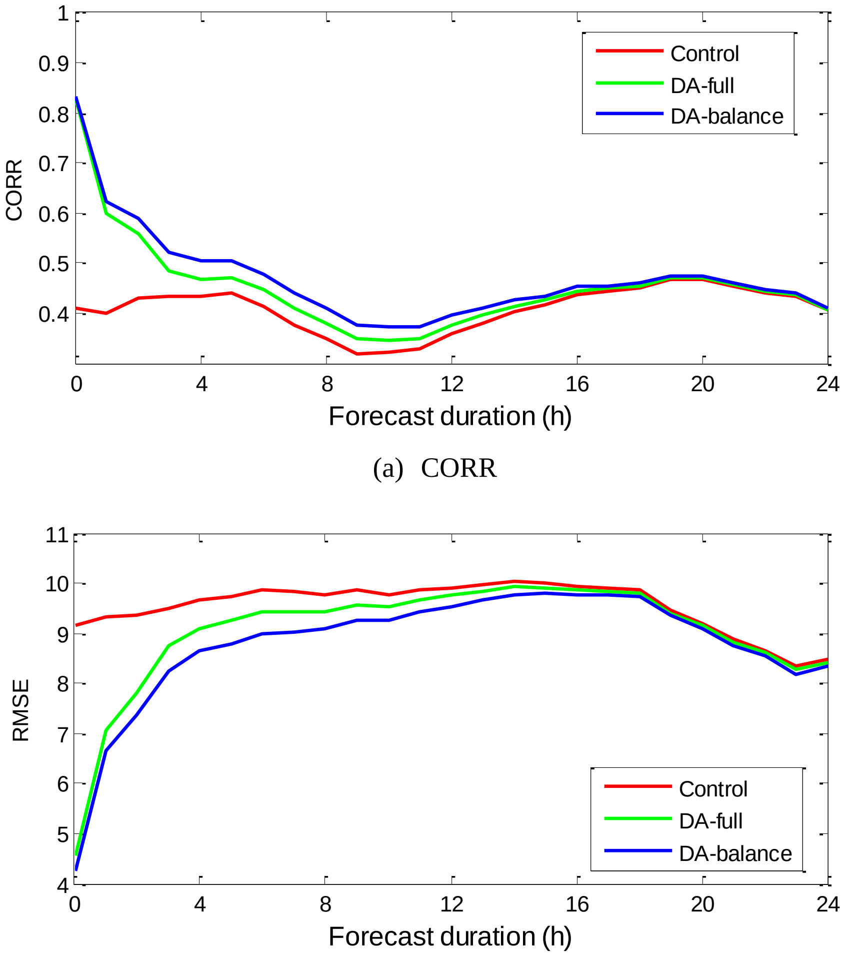
(a) Control

(b) DA-full

(c) DA-balance

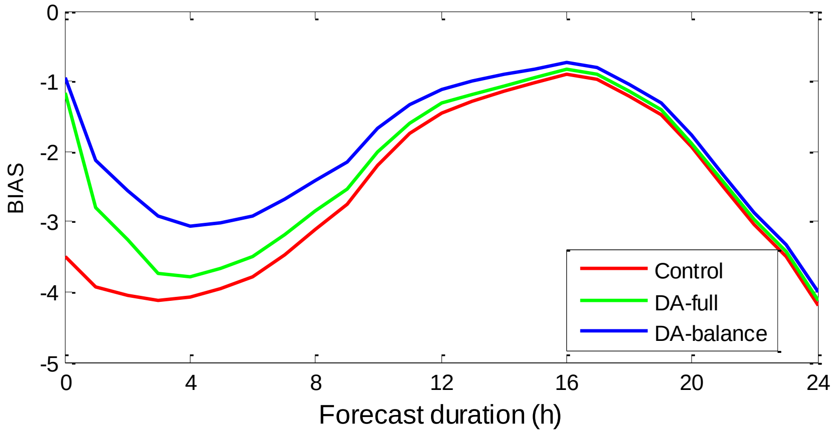
Figure 11. Scatter plots of observed concentrations of $PM_{2.5}$ versus simulated $PM_{2.5}$ concentrations of the experiments of (a) Control, (b) DA-full, and (c) DA-balance for all nine cases.

To evaluate the effects of the data assimilation, the CORR, RMSE and BIAS during the forecast time are calculated for each case, and their averaged results are showed in Figure 12. The CORRs of the DA-balance and DA-full experiments are very close (Fig. 12a). But, the difference increase after the first hour with a higher CORR in the DA-balance experiment. The CORR of the DA-balance experiment is substantially higher than that of the DA-full experiment from the 2nd hour to the 16th hour. Similar improvements for the RMSE and the BIAS of the DA-balance experiment are observed in Fig. 12b and Fig. 12c, respectively. The improvement for the BIAS in the DA-balance experiment is the most significant among these three statistical measures. The peak value of the improvement for the BIAS (Fig 12c) is at the 4th hour, and the improvement is distinct until the end of forecasts. These improvements indicate that the balance constraint is positive for the subsequent forecasts, which derives from the balanced initial distribution among species.



(a) CORR

(b) RMSE



(c) BIAS

Figure 12. The averaged (a) Correlations, (b) root-mean-square errors (RMSE in μg/m^3) and (c) mean bias (BIAS in μg/m^3) of the $PM_{2.5}$ concentration forecasts against observations as a function of forecast duration.

6. Summary and discussion

We examined the BEC in a 3DVAR system, which uses five control variables (EC, OC, NO_3, SO_4 and OTR) that are derived from the MOSAIC aerosol scheme in the WRF/Chem model. Based on the NMC method, differences within a month-long period between 24- and 48-h forecasts that are valid at the same time were employed in the estimation and analyses of the BEC. The background errors of these five control variables are highly correlated. Especially between EC and OC, their correlation is as large as 0.9.

A set of balance constraints was developed using a regression technique and incorporated in the BEC to account for the large cross correlations. We employ the the balance constraint to seperate the original full variables into balanced and unbalanced parts. The regression technique is used to express the balanced parts by the unbalanced parts. These unbalanced parts can be assumed independent. Then, the unbalanced parts are employed as control variables in the BEC statics. Accordingly, the standard deviations of these unbalanced variables are less than the standard deviations of the original variables. The horizontal correlation scales of unbalanced variables are closer than that of full variables on the effect of the balance constraints. And the vertical correlations of unbalanced variables show similar trend.

To evaluate the impact of the balance constraints on the analyses and forecasts, three groups of experiments, including a control experiment without data assimilation and two data assimilation experiments with and without balance constraints (DA-full and DA-balance), were performed. In the data assimilation experiments, the observations of surface $PM_{2.5}$ concentration and aircraft-speciated concentration of OC, NO_3 and SO_4 were assimilated. The observations of these three variables can spread to the two remaining variables in the increments of the DA-balance, which results in a more complex distribution. The evaluations of CORR, RMSE and BIAS for the initial analysis fields show more improvement in the DA-balance experiments, compared with the DA-full experiments. Though, these improvement are some slight. An important reason is that the surface $PM_{2.5}$ observations are independent from the aircraft observations. If we evaluate the analysis fields by the species observation of aircraft, there may be more significant improvements in the DA-balance experiments.

While the improvements increase after the first forecasting hour in the DA-balance experiments, compared with forecasts of the DA-full experiments. The improvements persist to the end of forecasts, and are substantial from the 2nd hour to the 16th hour (Fig. 12). These results suggested that the balance constraints can serve an import role for continually improving the skill of sequent forecasts. Note that some aircraft data are relative few, and some flight tracks are not around Los Angeles in some cases (Fig. 8). If there are more aircraft observations, the improvements of the DA-balance experiments should be more significant and durable.

The developed method for incorporating balance constraints in aerosol data assimilation can be employed in other areas or other applications for different aerosol models. For the aerosol variables in different models, some cross-correlations between different species or size bins should exist because their common emissions and diffusion processes are controlled by the same atmospheric circulation. Although these cross-correlations may be stronger than the cross-correlations of atmospheric or oceanic model variables, theoretic balance constraints, such as geostrophic balance or temperature-salinity balance, do not exist. We expected to discover a universal balance constraint that can describe the physical or chemical balanced relationship of aerosol variables, and utilize it in the data assimilation system. In addition, we expected to expand the balance constraint to include gaseous pollutants, such as nitrite (NO_2), sulfur dioxide (SO_2), and (carbon monoxide) CO. These gaseous pollutants are correlated with some aerosol species, such as NO_3, SO_4 and EC, which can improve the data assimilation analysis fields of aerosols by assimilating these gaseous observations. The assimilation of aerosol observations may improve the analysis fields of gaseous pollutants.

Code availablity

This data assimilation system is established by ourself. The code of this system can be obtained on request from the first author (zzlqxxy@163.com).

Acknowledgements

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Table 1 Regression coefficients of balance operator K and the coefficient of determination (regression coefficients correspond to ρ_{ij} in Eq. (7))

species	regression coefficient (ρ)					coefficient of determination (R^2)
EC	1					/
OC	0.90	1				0.86
NO_3	4.01	3.76	1			0.32
SO_4	1.35	-0.21	-3.15	1		0.48
OTR	2.93	2.35	0.28	0.60	1	0.96

Table 2 The periods of flight during CalNex 2010 and the initial time of assimilation

Number of cases	Start time of flight	End time of flight	Initial time of assimilation
1	18:00 UTC, May 16	01:42 UTC, May 17	00:00 UTC, May 17
2	17:28 UTC, May 19	00:10 UTC, May 20	18:00 UTC, May 19
3	17:28 UTC, May 21	00:10 UTC, May 21	18:00 UTC, May 21
4	23:08 UTC, May 24	05:23 UTC, May 25	00:00 UTC, May 25
5	01:59 UTC, May 30	07:45 UTC, May 30	06:00 UTC, May 30
6	05:00 UTC, May 31	10:54 UTC, May 31	06:00 UTC, May 31
7	07:59 UTC, June 2	14:09 UTC, June 2	12:00 UTC, June 2
8	07:59 UTC, June 3	14:041 UTC, June 3	12:00 UTC, June 3
9	17:56 UTC, June 14	23:35 UTC, June 14	18:00 UTC, June 14

Figure 1 Geographical display of the three-nested model domains. The innermost domain covers the Los Angeles basin; the black point denotes the location of Los Angeles.

Figure 2 Cross-correlations between emission species of E_EC, E_ORG, E_NO3, E_SO4 and E_PM25. The emission species data are derived from the NEI’05 emissions set for the innermost domain of the WRF/Chem model

Figure 3 Cross-correlations between the five variables of the BEC. These variables are (a) full variables and (b) unbalanced variables of EC, OC, NO_3, SO_4 and OTR.

Figure 4 Vertical profiles of the standard deviation of the variables. (a) full variables and (b) unbalanced variables

Figure 5 Same as Figure 4, with the exception of the horizontal auto-correlation curves of the variables. The horizontal thin line is the reference line of $e^{-\frac{1}{2}}(\approx 0.61)$ for determining the horizontal correlation scales.

Figure 6 Vertical correlations of the five variables of the BEC. The left column represents the full variables, and the right column represents the unbalanced variables.

Figure 7 The topography of the innermost domain and the locations of surface monitoring stations (black dots). The red square is the location of Los Angeles

Figure 8 Aircraft flight tracks during the time window of data assimilation for nine cases. The color of the track indicates the aircraft height.

Figure 9 Surface distributions of increments of the five variables of EC, OC, NO_3, SO_4 and OTR at 12:00 UTC on June 3, 2010. The left column and right column are from DA-full and DA-balance, respectively.

Figure 10 Same as Figure 9, with the exception of the vertical sections along 35 N.

Figure 11 Scatter plots of observed concentrations of $PM_{2.5}$ versus simulated $PM_{2.5}$ concentrations of the experiments of (a) Control, (b) DA-full, and (c) DA-balance for all nine cases.

Figure 12 The averaged (a) Correlations, (b) root-mean-square errors (RMSE in μg/m^3) and (c) mean bias (BIAS in μg/m^3) of the $PM_{2.5}$ concentration forecasts against observations as a function of forecast duration.