

Reply to the Comments by Referee #1 for Manuscript acpd-15-11573-2015

We appreciate the positive comments by the Referee #1 along with many valuable suggestions, which helped us improve the manuscript significantly. In the following, we have provided an item-by-item reply to the referee's comments.

General comments:

1. The fact that only one assimilation cycle is used reduced the scientific impact of this study. The authors argue that OMI data are only available in the morning so only one DA cycle was done, on 3 September in the morning. The period of the Nabi cyclone is between 29 August and 8 September, 2005. The switch of OMI from normal to zoom mode occurred on September 2 where no OMI data were available over the TC region for that day. If I understand well, OMI data is then available for the other days during the Nabi cyclone but only in the morning (around 4 UTC).

To me, the entire period of the Nabi cyclone could be addressed. Cycles without OMI data (between 6 and 24 UTC) might be replaced by a forecast run. In this way, the performance of DA experiment could be evaluated by measuring the skill of the system to forecast OMI data of the next days. This is one question to which the current state of the study does not answer: is DA of OMI data improve the forecast of the cyclone. Also, increasing the number of cycle would reinforce the results of section 3.4.

⇒ *We greatly appreciate the suggestions by the Referee #1, and agree that performance of data assimilation (DA) cycling with several cycles could make the DA more powerful. Although one can potentially have 4 cycles with a 6-hour assimilation window in a day, the limited availability of OMI observations over the model domain allows only one DA cycle per day. This is an extremely unfavorable situation for DA. Therefore, we only conducted the first DA cycle, which has the strongest impact among the cycles. We believe that our current single cycle DA experiment is sufficient to illustrate the effect of coupled meteorology-chemistry DA and demonstrate its potential.*

*A meaningful cycling of DA is inherently related to the **prediction component** of DA, as every new cycle begins from the forecast guess from the previous cycle. However, the **analysis component** of DA is also important, as it provides the impact of observations on the analysis produced by DA. **In the current research, we focus on the analysis component of DA, as the first step towards the eventual DA system for OMI observations.** We believe that there are sufficient new results in the context of the analysis component, which are relevant for coupled DA presented in this work. We plan to address some important issues related to the prediction component in the future studies.*

Although including the prediction component (e.g., cycling) is desirable, it has, unfortunately, several difficult aspects that are not possible to resolve in the current setup. It is known that realistic DA, including ours, is not perfect in providing dynamically balanced initial conditions, typically resulting in a forecast spin-up period where some of the analysis adjustments are filtered out. A practical remedy is to produce an improved fit to observations, bringing about the related stronger impact on dynamical model variables such as wind, temperature and pressure, which would eventually result in a longer, sustained impact into the forecast. However, given that the assimilation of OMI observations produces a stronger impact on chemical variables

and some but insufficient impact on dynamical initial conditions, the 24-hour forecast that we need for the next cycle would not be strongly influenced by the OMI observations. Thus we need to assimilate additional observations. Unfortunately, for typhoon this implies the need for assimilating satellite data, which are currently not available for the employed DA system, and thus would require additional development that is outside of the scope of this paper and is planned for the future step.

2. In the paper the terminologies “atmosphere chemistry model” or “atmospheric and chemical variables” is used. The chemical composition is part of the atmosphere state so I would change these terminologies by, e.g., “circulation chemistry model” and “physical (or dynamical) and chemical variables”

⇒ *We agree to the referee’s suggestion and introduce adequate changes throughout the manuscript. Given that the chemical composition is part of the atmospheric states, we suggest to change “atmosphere-chemistry model” to “meteorology-chemistry model”, and “atmospheric and chemical variables” to “meteorological and chemical variables”. Here “meteorological” variables include both “physical” and “dynamical” variables.*

Technical corrections:

P11576-L7: I would not use the term blending to describe DA method because it is too subjective while DA methods are an objective way to use model, a priori and observation information, as well as their error covariances to produce an analysis. Please, update the sentence.

⇒ *We have changed this part from “by blending the model and observations ...” to “... by combining the information from the model and observations in a mathematically consistent manner ...”*

P11576-L10-13: This sentence is not very clear. They are many reasons to assimilate ozone which are reviewed in Lahoz et al. (2007) for the stratosphere. Please, clarify the sentence.

⇒ *We have rewritten this part as “Ozone (O_3) has a relatively long photochemical lifetime and high concentrations at high latitude and in the stratosphere, except during ozone hole conditions. It is a passive tracer at synoptic scale or smaller; thus variations of total column O_3 in space and time are a result of the atmospheric flow, and is highly correlated to many meteorological variables in the upper troposphere (Wu and Zou, 2008). Assimilation of O_3 has several motivations such as (Lahoz et al., 2007): 1) taking better account of stratospheric O_3 when assimilating satellite radiance data; 2) leading to better radiative forcing when used by the model radiation scheme; 3) providing useful dynamical information via the motion of O_3 in the atmosphere; and 4) improving the accuracy of UV index forecasting. Moreover ...”*

P11579-L11-17: Some readers will probably not know the locations of Saipan, Kyushu, South Korea and Hokkaido. Would it be possible to mark these locations in Figure 1?

⇒ *We have redrawn Fig. 1 by marking those locations in the revised manuscript.*

P11580-L17-19: “It contains ...” The description of the observation operator that transforms modelled Ozone volume mixing ratio to total column is very short. Can you add more information; in particular are the averaging kernels used in the observation operator?

⇒ Following the referee’s suggestion we described the observation operator in more detail in the revised manuscript (please see the newly added equations from Eq. (1) to (5)). Please note that we are not using averaging kernels, following standard practice in DA, since we rely on the (multivariate) ensemble forecast error covariance for processing the information from the observation and the prior.

P11582-L13: “(ii) 200 hPa (lower stratosphere)”. 200 hPa is usually in the upper troposphere lower stratosphere (UTLS) so I would replace lower stratosphere by upper troposphere lower stratosphere.

⇒ It is rewritten as “(ii) 200 hPa (upper troposphere/lower stratosphere; UTLS)”.

P11582-L18-19: “These are ...” This sentence is not clear in particular after “... and the control forecast ...” Please, clarify.

⇒ It is rewritten as “**These are estimated by taking the difference between the ensemble perturbation forecasts (total of 32) and the control forecast in the ensemble system (Zupanski, 2005; Zhang et al., 2013).**” For further clarification, we have also rewritten P11583-L1 as “**forecasts with corresponding initial conditions \mathbf{x}^0 (i.e., control forecast) and ensemble initial conditions \mathbf{x}_n^0 (i.e., ensemble forecasts)**”.

P11583-L25-26: “..., provided total ...” This latter part of the sentence lack of clarity. Please, rephrase.

⇒ It is rewritten as “..., **provided that total ...**”.

P11584-L16: Do you mean Fig. 4b (instead of d)?

⇒ Fig. 4d is right. Please note that Fig. 4d (i.e., analysis increment of O_3 at 200 hPa) is right beside Fig. 4a (i.e., analysis increment of O_3 at 850 hPa).

P11584-L19: I would replace “... while the correlation is mixed ...” by “... while no clear correlation is found for ...”

⇒ It is rewritten as “..., **while no clear correlation is found in other regions.**”

P11585-L10: The term “validation” is in general used when the analyses are “validated” w.r.t. independent observations. Here, it is more a verification. Please, update the title of Sect. 3.4.

⇒ It is now changed to “**3.4 Verification of O_3 data assimilation**” in the revised manuscript. We have also corrected the **Abstract** (P11575-L18) accordingly as “**The analysis results are verified using ...**”.

P11587-L2: “... at the time” The meaning of this sentence is not clear. Please, rephrase.

⇒ It is rewritten as “**We include only a single data assimilation cycle since the OMI observations are covering the model domain only once per day (i.e., 06:00 UTC), and no other observations are available at that time.**”

General Comments:

An ensemble-based data assimilation, the maximum likelihood ensemble filter (MLEF) is employed and interfaced with the WRF-Chem to investigate the impact of ozone (O₃) assimilation on the structure of a tropical cyclone (TC). The results show that the O₃ assimilation has a notable impact on the analyses of other chemical variables (e.g., NO₂ and SO₂) as well as O₃ itself, and atmospheric variables (e.g., wind, temperature and specific humidity), especially near the TC case considered.

Please indicate in some detail: a) How was the coupling between MLEF and WRF-Chem implemented?; b) Please highlight the impact of including/excluding MLEF had on final result; c) Please highlight where in the WRF-Chem package is ozone taken into account.

Apart from these minor issues, this is a well-written and presented ms and I recommend publication once the minor comments are addressed.

⇒ We appreciate the positive comments by the Referee #2 along with valuable suggestions. In the following, we have provided an item-by-item reply to the referee's comments.

a) How was the coupling between MLEF and WRF-Chem implemented?

⇒ The coupling between the MLEF and WRF-Chem is done through an interface module that transforms the MLEF control variables into a WRF-Chem `netcdf` file, and vice versa. This interface module is a component of MLEF, and thus the WRF-Chem is not altered.

b) Please highlight the impact of including/excluding MLEF had on final result.

⇒ The impact of including/excluding MLEF on the final result has been described in detail in Section 3. In overview, the ozone observations had an impact on ozone analysis, as expected. The important new impact of ozone observations, enabled through the use of ensemble-based forecast error covariance, includes changes in the initial conditions of dynamical variables, such as wind and temperature, and to some extent moisture. More specific impacts of including MLEF assimilation of O₃ are discussed in Figs. 4 and 5 in detail – changes of chemical variables in Fig. 4 and changes of atmospheric variables in Fig. 5, respectively.

c) Please highlight where in the WRF-Chem package is ozone taken into account.

⇒ The WRF-Chem chemistry package is chosen in the `namelist.input` as a standard option #6 (CBMZ). This chemistry package includes the prediction of ozone and several other chemical constituents. Since the control variable list in the MLEF includes the ozone, it makes possible adjustment of ozone initial conditions in data assimilation.

Impact Ensemble data assimilation of total column ozone observations using a coupled meteorology-chemistry model and its impact on the structure of a tropical cyclone using coupled atmosphere-chemistry data assimilation Typhoon Nabi (2005)

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Abstract. ~~Since the air quality forecast is related to both chemistry and meteorology, the coupled atmosphere-chemistry data assimilation (DA) system is essential to air quality forecasting.~~ Ozone (O₃) plays an important role in chemical reactions and is usually ~~assimilated in chemical DA~~ incorporated in chemical data assimilation (DA). In tropical cyclones (TCs), O₃ usually shows a lower concentration inside the eyewall and an elevated concentration around the eye, impacting ~~atmospheric meteorological~~ as well as chemical variables. To identify the impact of O₃ observations on TC structure, including ~~atmospheric-meteorological~~ and chemical information, we ~~employed-developed a coupled meteorology-chemistry DA system by employing~~ the Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) with-and an ensemble-based DA algorithm – the maximum likelihood ensemble filter (MLEF). For a TC case that occurred over ~~the~~ East Asia, Typhoon Nabi (2005), our results indicate that the ensemble forecast is reasonable, accompanied with larger background state uncertainty over the TC, and also over eastern China. Similarly, the assimilation of O₃ observations impacts ~~atmospheric-meteorological~~ and chemical variables near the TC and over eastern China. The strongest impact on air quality in the lower troposphere was over China, likely due to the pollution advection. In the vicinity of the TC, however, the strongest impact on chemical variables adjustment was at higher levels. The impact on ~~atmospheric-meteorological~~ variables was similar in both over China and near the TC. The analysis results are ~~validated-verified~~ using several measures that include the cost function, root-mean-squared (RMS) error with respect to observa-

tions, and degrees of freedom for signal (DFS). All measures indicate a positive impact of DA on the analysis – the cost function and ~~root-mean-square~~RMS error have decreased by 16.9 and 8.87 %, respectively. In particular, the DFS indicates a strong positive impact of observations in the TC area, with a weaker maximum over northeast China.

1 Introduction

The air quality forecast is related to emissions, transport, transformation and removal processes, and to the prevailing meteorological conditions. Therefore, the coupled ~~atmosphere–chemistry–meteorology–chemistry~~ model is essential for the air quality and weather forecasting (e.g., Carmichael et al., 2008). The coupled system forecast is improved through coupled ~~atmosphere–chemistry–meteorology–chemistry~~ data assimilation (DA), which estimates the best initial conditions by ~~blending the combining the information from the~~ model and observations in a mathematically consistent manner (e.g., Houtekamer and Mitchell, 1998; Elbern and Schmidt, 1999; Wang et al., 2001; Evensen, 2003; Park and Zupanski, 2003; Navon, 2009; Zupanski, 2009; Park et al., 2015).

Ozone (O_3) ~~is usually assimilated in a chemical DA because it represents the atmospheric flow as a passive tracer at synoptic or smaller scales and has a~~ relatively long photochemical lifetime and high concentrations at high latitude and in the stratosphere, except during ozone hole conditions; ~~and at high latitudes (e.g., Lahoz et al., 2007; Wu and Zou, 2008);~~ It is a passive tracer at synoptic scale or smaller; thus variations of total column O_3 in space and time are a result of the atmospheric flow, and is highly correlated to many meteorological variables in the upper troposphere (Wu and Zou, 2008). Assimilation of O_3 has several motivations such as (Lahoz et al., 2007; ~~Wu and Zou, 2008~~): 1) taking better account of stratospheric O_3 when assimilating satellite radiance data; 2) leading to better radiative forcing when used by the model radiation scheme; 3) providing useful dynamical information via the motion of O_3 in the atmosphere; and 4) improving the accuracy of UV index forecasting. Moreover, the improved stratospheric O_3 distribution by DA can affect ~~atmospheric–meteorological~~ variables such as stratospheric winds and temperature as well as other chemical variables (e.g., Lahoz et al., 2007; Park et al., 2015).

O_3 is also relevant to the structure of tropical cyclones (TCs), showing a lower concentration just inside the eyewall and elevated concentration around the eye (e.g., Carsey and Willoughby, 2005; Zou and Wu, 2005; Wu and Zou, 2008), which is caused by the updraft in the eyewall and subsidence in the eye (Zou and Wu, 2005). Using these relations, the daily total column O_3 from Total Ozone Mapping Spectrometer (TOMS) showed that mutual adjustment occurred between the TC and its upper tropospheric environment on a synoptical timescale (Rodgers et al., 1990; Stout and Rodgers, 1992). The linear relationship between total column O_3 from TOMS and mean vertically-integrated potential vorticity (MPV) was used to improve hurricane or winter storm prediction (e.g., Jang et al., 2003; ~~Wu and Zou, 2008; Zou~~ Zou and Wu, 2005; Wu and Zou, 2008). However, these

studies employed ~~an-atmospheric-a meteorological~~ model, not the coupled ~~atmosphere-chemistry~~
55 ~~meteorology-chemistry~~ model. They used the standard dynamical variables as control variables and empirical regressions to develop a cross-correlation between O_3 and dynamical model variables.

In this study, we directly assimilate the total column O_3 from the Ozone Monitoring Instrument (OMI) to identify the impact of O_3 observations on TC structure including ~~atmospheric-meteorological~~ and chemical information in a coupled ~~atmosphere-chemistry-meteorology-chemistry~~ model (e.g.,
60 WRF-Chem) with ensemble-based DA system (e.g., Maximum Likelihood Ensemble Filter; MLEF). We define an augmented control variable that ~~includes both dynamical~~ contains both meteorological and chemical variables. Here meteorological variables consist of dynamical variables (e.g., wind components) and physical variables (e.g., water vapor, cloud water, etc.). Therefore, the cross-correlations between ~~dynamical-meteorological~~ and chemical variables are obtained directly from
65 ensemble forecasts ~~-(e.g., Park et al., 2015).~~ Section 2 describes the methodology, and Sect. 3 presents results. Conclusions are provided in Sect. 4.

2 Methodology

2.1 Model

In this research, we use the Weather Research and Forecasting (WRF) model coupled with Chem-
70 istry (WRF-Chem) version 3.4.1 as a prediction model on a regional scale. It simulates the emission, transport, mixing and chemical transformation of trace gases and aerosols simultaneously with meteorology (Grell et al., 2005). The WRF-Chem uses configuration options for various ~~atmospheric~~ meteorological processes such as the WRF Single-Moment 6-class (WSM6) scheme for the microphysics, the Community Atmospheric Model (CAM) scheme for the radiation physics, the Monin-
75 Obukhov scheme for the surface layer, the Noah land surface model for the land surface, the Yonsei University (YSU) scheme for the planetary boundary layer, and the Kain-Fritsch scheme for the cumulus parameterization. These are the recommended physics options for the regional climate case at 10–30 km grid size. As an advection option, the monotonic transport is applied to turbulent kinetic energy and scalars such as mixing ratios of water vapor, cloud water, rain, snow and ice and chem-
80 ical species, which is commonly used for real-time and research applications (e.g., Chapman et al., 2009; Yang et al., 2011). Regarding the chemical mechanism, the Carbon Bond Mechanism version Z (~~CBMZ~~CBM-Z) without Dimethylsulfide scheme is used for gas-phase chemistry. The CBM-Z includes the prediction of O_3 and several other chemical constituents (Fast et al., 2006).

In terms of the DA system, we use an ensemble-based DA method called the Maximum Like-
85 lihood Ensemble Filter (MLEF; Zupanski, 2005; Zupanski et al., 2008). The MLEF generates the analysis solution which maximizes the likelihood of the posterior probability distribution, obtained by minimization of a cost function. The MLEF belongs to the family of deterministic ensemble filters, hence it is a hybrid between variational and ensemble DA methods. The MLEF employs a cost

function derived using a Gaussian probability density function and produces both the analysis and the background error covariance (Zupanski, 2005). It is well suited for use with highly nonlinear observation operators, for a small additional computational cost of minimization using the Hessian preconditioning (Zupanski, 2005; Zupanski et al., 2007, 2008), and has been employed in many studies including uncertainty analysis, parameter estimation and data assimilation (e.g., Zupanski and Zupanski, 2006; Zupanski et al., 2007a; Lokupitiya et al., 2008; Kim et al., 2010; Apodaca et al., 2014; Tran et al., 2014; Park et al., 2015).

The coupling between the MLEF and WRF-Chem is made through an interface module that transforms the MLEF control variables into the netcdf file of WRF-Chem, and vice versa. This interface module is a component of MLEF, and hence the WRF-Chem is not altered.

2.2 Observations

Satellite retrievals often provide estimates of chemical concentration as a total vertical column, and they cover a wide geographical range compared to other measurements (e.g., Silver et al., 2013). In our study, the total column O_3 obtained by OMI is used as an observation. OMI is a nadir-viewing near-UV/Visible CCD spectrometer aboard NASA's Aura satellite (OMI Team, 2012). The total column O_3 is Level 2 data (OMTO3) based on the Total Ozone Mapping Spectrometer (TOMS) v8.5 algorithm, which is obtained from an orbital swath with a resolution of $13\text{ km} \times 24\text{ km}$ at nadir (OMI Team, 2012). It achieves global coverage in one day. In this experiment, we did not apply the quality flags because the first appearance of the row anomaly that affects particular viewing directions, corresponding to the rows on the CCD detectors (OMI Team, 2012) did not occur in 2005, when the TC case considered occurred (i.e., Typhoon Nabi, 2005). Therefore, we employ the OMI data without quality flags.

Figure 1 shows the total column O_3 from OMI at 04:05 UTC 3 September 2005. It shows a lower concentration just inside the eyewall and elevated concentration around the eye. This distinct distribution is well described when the TC has the strongest intensity in the intensifying stages (e.g., Carsey and Willoughby, 2005). Note that OMI switches from its normal global mode to zoom-in mode, to perform spatial zoom (higher resolution) measurements, for a 24 h period about once a month. It occurs when OMI finishes its last orbital pass over Europe, and returns to global mode after 14–15 orbits or about 24 h later. During this period of zoom-in mode, OMI has no global coverage of data (OMI Team, 2012). Typhoon Nabi (2005) reached the maximum intensity on 2 September when OMI entered in the zoom-in mode. Due to the lack of ~~ozone~~- O_3 data in our domain on 2 September, we have alternatively chosen 3 September for the analysis of O_3 properties during the maximum development of the TC case.

2.3 Experimental design

For the TC case, we choose Typhoon Nabi (2005), which lasted several days from 29 August 2005 until 8 September 2005. Nabi moved westward after its formation and passed near Saipan on 31 August as an intensifying TC, transformed to a super typhoon on 1 September, and reached its peak with winds of 175 km h^{-1} (10-min average) on 2 September. It became weak while turning to the north and striking Kyushu on 6 September. Nabi turned to the northeast after passing by South Korea, and transformed to an extratropical cyclone passing over Hokkaido on 8 September.

In general the DA is composed of two components – prediction and analysis. A meaningful cycling of DA is inherently related to the prediction component, as every new cycle begins from the forecast guess from the previous cycle. The analysis component of DA is also important, as it provides the impact of observations on the analysis produced by DA. In the current research, we focus on the analysis component of DA, as the first step towards the eventual DA system for OMI observations.

Conducting the DA cycling with several cycles can make DA more powerful. Although one can potentially have 4 cycles with a 6-hour assimilation window in a day, the limited availability of OMI observations over the model domain allows only one DA cycle per day. Therefore, we only perform the first DA cycle, which has the strongest impact among the cycles. We believe that this single cycle DA experiment is sufficient to illustrate the effect of coupled meteorology-chemistry DA.

We focused on a single DA cycle from 00:00 to 06:00 UTC 3 September 2005, which is one of the strongest periods of its lifetime. We conduct the experiment with 32 ensembles and 6 h assimilation window. Note that the OMI observations have an approximate frequency of once per day over the typhoon and the surrounding geographical area. Therefore, adding more ~~data-assimilation-DA~~ cycles would not be beneficial since no additional data are available. In the future we plan to add a -capability to assimilate other observations, such as ~~atmospheric-meteorological~~ observations and all-sky infrared radiances from a -geostationary satellite.

The initial and lateral boundary conditions for ~~atmospheric-meteorological~~ states are provided by the National Centers for Environmental Prediction (NCEP) Global Forecasting System (GFS), while those for chemical variables are obtained from the Model for Ozone and Related chemical Tracers (MOZART) chemistry global model of the National Center for Atmospheric Research (NCAR)/Atmospheric Chemistry Division (ACD). The WRF-Chem is set up with a horizontal resolution of 30 km and 51 vertical levels with the bottom at the ground and the top at 10 hPa using a terrain-following hydrostatic pressure coordinate (Skamarock et al., 2008).

The model domain is centered over the Korean Peninsula, covering an area of approximately $3900 \text{ km} \times 4400 \text{ km}$ with 132×147 horizontal grid points. The control variables defined in the coupled ~~atmosphere-chemistry-meteorology-chemistry~~ DA are the WRF-Chem prognostic variables that contain ~~dynamical-meteorological~~ variables such as winds, perturbation potential temperature, perturbation geopotential, water vapor mixing ratio and perturbation dry air mass in a column, and

the chemical variables such as ozone (O_3), nitrates (NO , NO_2 , NO_3), and sulfur dioxide (SO_2). The experiments consist of (i) the forecast (without DA) which is useful to understand the synoptic situation and background error covariance, and (ii) the analysis (with DA) which is useful to understand the assimilation impacts.

2.4 Bias correction of total column O_3

We define the observation operator transforming the WRF-Chem O_3 forecast to the total column O_3 observation. It contains the calculation of total column O_3 ~~unit conversion from ppmv (parts per million by volume) to~~ with unit conversion and bi-linear interpolation, that is: 1) to transform the physical units of O_3 from the model-produced concentrations in parts-per-million-volume (ppmv) units to the OMI data in Dobson Units (DU) ~~and~~, and 2) to transform the O_3 amount from the model grid levels to vertically integrated value at the ~~bi-linear interpolation to the~~ observation location. ~~The~~ Mathematically, the operator can be written as

$$h(x) = h_i h_c h_u(x) \quad (1)$$

where x denotes an input model variable (e.g., concentration), and h_i represents the horizontal interpolation operator, h_c the vertical column integration and h_u the unit transformation from ppmv to DU. The unit transformation for ozone, h_u , is given by

$$h_u(x) = \frac{A \cdot 10^{-8} \Delta p}{g \cdot m_d} \cdot x \quad (2)$$

where $A = 6.02252 \times 10^{23}$ is the Avogadro number, Δp is the vertical increment of pressure in the layer (hPa), g is the gravity constant, and m_d is the molecular weight of dry air (kg/mol). The vertical column integration, h_c , is simply

$$h_c(s) = \sum_{k=1}^K s_k \quad (3)$$

where s_k is the ozone in DU at layer k , and K denotes the number of vertical layers. Finally the bi-linear horizontal interpolation, h_i , is

$$h_i(r) = \sum_{i=1}^I w_i r_i \quad (4)$$

where r_i is the vertically-integrated ozone at grid point i , w_i is the bi-linear observation weights at grid point i , and I denotes the number of grid points used in interpolation ($I = 4$ in our case). After combining (2)-(4) into (1), the observation operator for OMI observations becomes

$$h(x) = \sum_{i=1}^I w_i \left(\sum_{k=1}^K \frac{A \cdot 10^{-8} (\Delta p)_k}{g \cdot m_d} \cdot x_k \right)_i \quad (5)$$

In these processes, the most demanding part of the observation operator is bias correction of total column O_3 observation. Although we use the reference pressure at the model top as 10 hPa, which is the highest value we could use in the current model version, there are still considerable amounts of O_3 in the stratosphere that could not be included in the calculation of the model guess (e.g., background). Since this creates a negative bias in the mean observation error, we introduce a multiplicative bias correction ε to preserve positive-definiteness of the bias-corrected guess (Apodaca et al., 2014) as

$$h_B(\mathbf{x}) = \varepsilon \cdot h(\mathbf{x}) \quad (6)$$

where \mathbf{x} is the model state vector. With the multiplicative bias correction in Eq. (46), we can make a new cost function in unbiased form as

$$J(\mathbf{x}) = \frac{1}{2}(\mathbf{x} - \mathbf{x}_b)^T \mathbf{P}_f^{-1}(\mathbf{x} - \mathbf{x}_b) + \frac{1}{2}[\mathbf{y} - h_B(\mathbf{x})]^T \mathbf{R}^{-1}[\mathbf{y} - h_B(\mathbf{x})] \quad (7)$$

where \mathbf{x} is the model state vector, \mathbf{x}_b is the prior (background) state, \mathbf{y} is the observation vector, and the superscript T means a transpose. Here, h is the nonlinear observation operator, \mathbf{P}_f is the background (forecast) error covariance matrix in the ensemble subspace, and \mathbf{R} is the observation error covariance matrix. Equation (27) is the cost function used in DA, provided ε can be estimated. The optimal value of parameter ε is obtained by implicitly assuming lognormal probability density function errors for a multiplicative bias correction in Eq. (46) (e.g., Apodaca et al., 2014) as

$$\varepsilon = \varepsilon_0 \exp \left[\frac{\frac{1}{N} \sum_{i=1}^N \log \left(\frac{\mathbf{y}_i}{\varepsilon_0 h(\mathbf{x})_i} \right)}{1 + \frac{r_0}{w_0}} \right] \quad (8)$$

where ε_0 is a guess parameter value and N is the number of observations. The empirical weighting values are set to $r_0 = w_0 = 0.5$ which implies having the same confidence in observations and the guess. We assume the starting value of the bias to be

$$\varepsilon_0 = \frac{\bar{\mathbf{y}}}{\overline{h(\mathbf{x})}} \quad \text{where} \quad \bar{\mathbf{y}} = \frac{1}{N} \sum_{i=1}^N \mathbf{y}_i, \quad \overline{h(\mathbf{x})} = \frac{1}{N} \sum_{i=1}^N h(\mathbf{x})_i \quad (9)$$

Equation (38) is calculated once in every DA cycle.

3 Results

A specific characteristic of our experiments is that both atmospheric-meteorological and chemical variables are used as control variables in DA. Regarding the atmospheric-meteorological variables, we focus on what is related to the TC formation and development, such as the temperature, wind, and water vapor. Regarding the chemical variables, we select the chemical constituents such as O_3 , NO_2 and SO_2 . These are used to identify the impact of O_3 observations on the TC structure in a WRF-Chem-MLEF system.

3.1 Synoptic situation with ensemble WRF-Chem forecast

In general, observations show that SO₂ has larger concentrations in the troposphere while O₃ and NO₂ have larger concentrations in the stratosphere (e.g., Meena et al., 2006). However, in East Asia, especially in eastern China, there is a significant tropospheric NO₂ concentration because of the industrialized and urbanized part of China (Richter et al., 2005; Ohara et al., 2007). Regarding the ~~atmospheric~~-meteorological variables, temperature and water vapor have higher values in the troposphere, while wind has larger speed near the tropopause. To consider these characteristics, we focused on two pressure levels: (i) 850 hPa (lower troposphere) and (ii) 200 hPa (~~lower~~ stratosphereupper troposphere/lower stratosphere; UTLS). Our ensemble WRF-Chem forecast also supports these general distributions of control variables, which are not shown in this paper.

3.2 Background error covariance

The background error covariance represents the background state uncertainty (e.g., Zupanski and Zupanski, 2007; Kim et al., 2010). These are estimated by taking the difference between ~~each of~~ the 32 ensemble members-the ensemble perturbation forecasts (total of 32) and the control forecast in the ensemble system (~~Zhang et~~ Zupanski, 2005; Zhang et al., 2013). In our study, the ensemble WRF-Chem-MLEF estimates the background error covariance defined in Zupanski (2005) as

$$\mathbf{P}_f = \mathbf{P}_f^{1/2} \left(\mathbf{P}_f^{1/2} \right)^T, \quad \mathbf{P}_f^{1/2} = (p_1^f \cdots p_N^f), \quad p_n^f = m(\mathbf{x}_{0n}^{n0}) - m(\mathbf{x}_0^0) \quad (10)$$

where the index n is an ensemble member, N is the total number of ensemble forecasts, m is the WRF-Chem model, and the subscript 0 denotes the initial time of the forecast with corresponding initial conditions ~~\mathbf{x}_0~~ - \mathbf{x}_0^0 (i.e., control forecast) and ensemble initial conditions ~~\mathbf{x}_0^n~~ - \mathbf{x}_n^0 (i.e., ensemble forecasts). In this experiment, the initial ensemble perturbations are generated by using the lagged forecast outputs (Zhang et al., 2013).

Being calculated from the WRF-Chem ensemble forecast, the flow-dependent background error covariance is defined for ~~atmospheric~~-meteorological and chemical variables, which allows chemistry observations to impact ~~atmospheric~~-meteorological variables in DA. In Zhang et al. (2013), a larger background state uncertainty was found in the storm region. Our results also identify the larger background state uncertainty near the TC, similar to Kim et al. (2010). Figure 2 shows the standard deviation (SD) of background error covariance for chemical variables. O₃ in particular (Fig. 2a and d, respectively) shows a large background state uncertainty near the TC, with the maximum of 0.024 ppmv at 200 hPa (Fig. 2d). The background state uncertainties of NO₂ and SO₂ at 200 hPa (Fig. 2e and f, respectively) are located near the TC, characterized by small magnitude and weak influence on tropospheric pollution. On the other hand, the background state uncertainties of NO₂ and SO₂ at 850 hPa (Fig. 2b and c, respectively) have more impact on central eastern China, implying no visible (or obvious) impact of the low-level NO₂ and SO₂ on the TC.

The SD of background error covariance for ~~atmospheric-meteorological~~ variables appear to be more related to the TC structure (see Fig. 3). In particular, wind (Fig. 3a and d, respectively) shows a larger background state uncertainty near the TC at both pressure level, especially in the eye region at 850 hPa (Fig. 3a). Temperature (Fig. 3b and e, respectively) also shows a larger background state uncertainty near the TC, especially at 200 hPa (Fig. 3d). Regarding the water vapor mixing ratio (Fig. 3c and f, respectively), there is a larger background state uncertainty in the eye region at both pressure levels. Larger background state uncertainty potentially implies a stronger analysis correction, provided that total column O₃ observations are available.

3.3 Analysis increment through the O₃ data assimilation

We assess the impact of the assimilated O₃ observations using analysis increments ($\mathbf{x}_a - \mathbf{x}_b$), which show the correction of the background state using the observations. It is calculated by the following variable transformation (Zhang et al., 2013; Zupanski, 2005)

$$\mathbf{x}_a - \mathbf{x}_b = \mathbf{P}_f^{1/2} \{ \mathbf{I} + [\mathbf{Z}(\mathbf{x}_b)]^T \mathbf{Z}(\mathbf{x}_b) \}^{-1/2} \zeta \quad (11)$$

where ζ is the control variable in the ensemble space; the matrix in Eq. (611) is equal to the inverse of the square root Hessian of the cost function in Eq. (27); \mathbf{Z} is the observation information matrix with column vectors $\mathbf{z}_i = \mathbf{R}^{-1/2} [h(\mathbf{x}_i) - h(\mathbf{x}_b)]$, where the index i denotes the ensemble member).

Figure 4 shows the analysis increments ($\mathbf{x}_a - \mathbf{x}_b$) of chemical variables obtained by assimilating O₃ observations. By comparing Figs. 2 and 4 one can notice that the O₃ analysis increments are in agreement with background state uncertainties, as expected from Eq. (611). At 850 hPa, the O₃ analysis increment has an increase near the TC, but a decrease over China (Fig. 4a). At 200 hPa, however, there is an increase of O₃ near the TC, and marginal change over China (Fig. 4d). The strong positive response has the largest value of approximately 0.024 ppmv. At 200 hPa, positive O₃ analysis increments are correlated with positive NO₂ (Fig. 4e) and SO₂ (Fig. 4f) increments in the TC region, while ~~the correlation is mixed for~~ no clear correlation is found in other regions. ~~While~~ Note that NO₂ and SO₂ are not related to the TC at 850 hPa, ~~while~~ while the O₃ analysis increments are correlated with NO₂ (Fig. 4b) and SO₂ (Fig. 4c), increasing in central eastern China and Korea and decreasing in northeastern China.

Figure 5 shows the analysis increments ($\mathbf{x}_a - \mathbf{x}_b$) of ~~atmospheric-meteorological~~ variables by O₃ assimilation. Corresponding to background state uncertainties, the analysis increments of wind show notable impact on both lower and upper pressure levels. Positive O₃ increments correspond to positive wind increments at 850 hPa (Fig. 5a), especially in the eye region, and to positive wind increments at 200 hPa (Fig. 5d) in the TC and in the northeastern China and Korea. Regarding the temperature impact (Fig. 5b and e, respectively), the positive O₃ increments generate temperature cooling near the TC and warming over northeastern China. Regarding the water vapor mixing ratio, positive O₃ increments generate a reduction of water vapor mixing ratio (Fig. 5c and

f, respectively) near the TC as well as in the eye region at both pressure levels. At 850 hPa, the water vapor mixing ratio is increasing with positive O_3 increments over the northeastern China (Fig. 5c). These results illustrate that chemical observations can impact not only the chemical variables but also the ~~atmospheric-meteorological~~ variables, due to using the ensemble-based coupled ~~atmosphere-chemistry-meteorology-chemistry~~ background error covariance, ~~as indicated by Park et al. (2015).~~

3.4 ~~Validation-Verification~~ of O_3 data assimilation

As a verification measure, we examine the O_3 assimilation impact on the cost function and on the root mean square (RMS) error with respect to O_3 observations, the same data used in the analysis.

The cost function of O_3 driven by Eq. (27) has decreased from 0.36924×10^4 (background) to 0.30689×10^4 (analysis), i.e., it is reduced by approximately 16.9 %. The RMS error, calculated as

$$RMS_a = \sqrt{\frac{1}{N} \sum [y - h(x_a)]^2}, \quad RMS_b = \sqrt{\frac{1}{N} \sum [y - h(x_b)]^2} \quad (12)$$

~~where subscripts a and b denotes analysis and background, respectively,~~ has also decreased from 0.16684×10^2 DU (background) to 0.15204×10^2 DU (analysis), i.e., by about 8.87 %. These results suggest that O_3 assimilation has produced a significant improvement in the initial conditions.

In addition, the impact of ~~total-column-total column~~ O_3 observations is also quantified in terms of the uncertainty reduction. With the Gaussian probability assumption, the information content of observations can be represented as the degrees of freedom for signal (~~d_s~~) (e.g., DFS; Rodgers, 2000) ~~as, d_s , as~~

$$d_s = \text{tr} [\mathbf{I} - \mathbf{P}_a \mathbf{P}_f^{-1}] \quad (13)$$

where tr is trace functions, \mathbf{I} is the identity matrix, and \mathbf{P}_a and \mathbf{P}_f are the analysis and background error covariances. Here d_s can also be expressed as

$$d_s = \sum_i \frac{\lambda_i^2}{1 + \lambda_i^2} \quad (14)$$

where λ_i are the eigenvalues of the observation information matrix (e.g., Zupanski et al., 20072007b).

Note from Eq. (914) that the d_s are strictly a non-negative measure: zero values indicate no impact of observations, while positive values indicate a reduction of uncertainty due to assimilation. As shown in Zupanski et al. (20072007b), the estimation of Eq. (914) is also useful in a reduced-rank setting of ensemble ~~data-assimilationDA~~.

~~In Fig. Figure 6 we show the degree of freedom for signal of the~~ ~~shows the DFS of~~ assimilated total column O_3 observations. One can note that it generally coincides with the satellite path, and thus with observations, as expected. The area with the maximum impact is near the TC location, indicating that it is the area where the total column O_3 observation had the strongest impact. In agreement with the analysis increments, there exists a secondary maximum over northeast China, and

a smaller one over the Yellow Sea. Given that the DA system includes ~~atmospheric-meteorological~~ and chemical control variables, this result also indicates that O₃ total column observations have a positive impact on both the ~~atmospheric-meteorological~~ and chemical components of the WRF-Chem system, especially in the TC area.

4 Conclusions

In this study, we investigated the impact of ozone (O₃) assimilation on the structure of a tropical cyclone (TC). We directly assimilated the total column O₃ from the Ozone Monitoring Instrument (OMI) in a coupled ~~atmosphere-chemistry-meteorology-chemistry~~ modelling system – the Weather Research and Forecasting (WRF) model coupled with Chemistry (WRF-Chem). An ensemble-based data assimilation (DA) method, the maximum likelihood ensemble filter (MLEF), is employed and interfaced with the WRF-Chem. We include only a single ~~data-assimilation-DA~~ cycle since the OMI observations are covering the model domain only once per day ~~-(i.e., 06:00 UTC)~~, and no other observations ~~were available at the time. The use of a single data assimilation cycle limits the conclusions that can be drawn regarding the robustness of the data assimilation system, but it does not impact the performance and implications of using a coupled atmosphere-chemistry data assimilation system. are available at that time.~~

Our results show that the O₃ assimilation has a notable impact on the analyses of other chemical variables (e.g., NO₂ and SO₂) as well as O₃ itself, and ~~atmospheric-meteorological~~ variables (e.g., wind, temperature~~and specific humidity, water vapor, etc.~~), especially near the TC case considered. These ~~atmospheric-meteorological~~ variables are closely related to the TC structure and other properties. The O₃ observations can affect other chemical and ~~dynamical-meteorological~~ variables, and thus the TC itself. For example, temperature is related to development, wind to intensity, and ~~specific humidity-water vapor~~ to precipitation of the TC. Therefore, the implied corrections of these variables in TC regions have a potential to improve the understanding, and eventually the forecast of TCs.

In our ~~data-assimilation-DA~~ experiments, the ensemble forecast error, given by the background error ~~SD~~standard deviation, appears reasonable with larger uncertainty over the TC area and also over eastern China. The ~~RMS-root mean square~~ error reduction indicates an improvement of the optimal analysis state, while the degrees of freedom for signal indicate a reduction of the uncertainty of the optimal analysis.

The use of a single DA cycle limits the conclusions that can be drawn regarding the robustness of the DA system, but it does not impact the performance and implications of using a coupled meteorology-chemistry DA system. It is desired to perform a DA cycling with multiple cycles (i.e., the prediction component of DA); however, it has several difficult aspects that are not possible to resolve in the current setup. It is known that the realistic DA is not perfect in providing dynamically balanced initial conditions, typically resulting in a forecast spin-up period where some of the analysis

adjustments are filtered out. A practical remedy is to produce an improved fit to observations, bringing about the related stronger impact on dynamical model variables (e.g., wind, temperature and pressure), which would eventually result in a longer, sustained influence into the forecast. However, given that the assimilation of OMI observations exerts a stronger impact on chemical variables than dynamical initial conditions, the 24-hour forecast that we need for the next cycle would not be strongly influenced by the OMI observations. Thus we need to assimilate additional observations.

As a future study, we plan to explore the longer ~~data-assimilation~~ DA periods (e.g., several days) to assess the impact of O_3 observation ~~impact~~ on the track, intensity and precipitation of TCs. Although we have only one available observation product per day for O_3 , we anticipate a positive impact of assimilation. In order to obtain more improved DA effects, in addition to O_3 , we plan to assimilate NO_2 and SO_2 observations, as well as ~~atmospheric~~ meteorological observations and all-sky infrared satellite radiances from a geostationary satellite that will be launched in the near future. Noting that NO_2 and SO_2 show high concentrations in East Asia, especially over eastern China, we expect to improve our understanding of the TC structure and the transboundary air pollution as well through assimilation of such chemical compositions from satellite observations.

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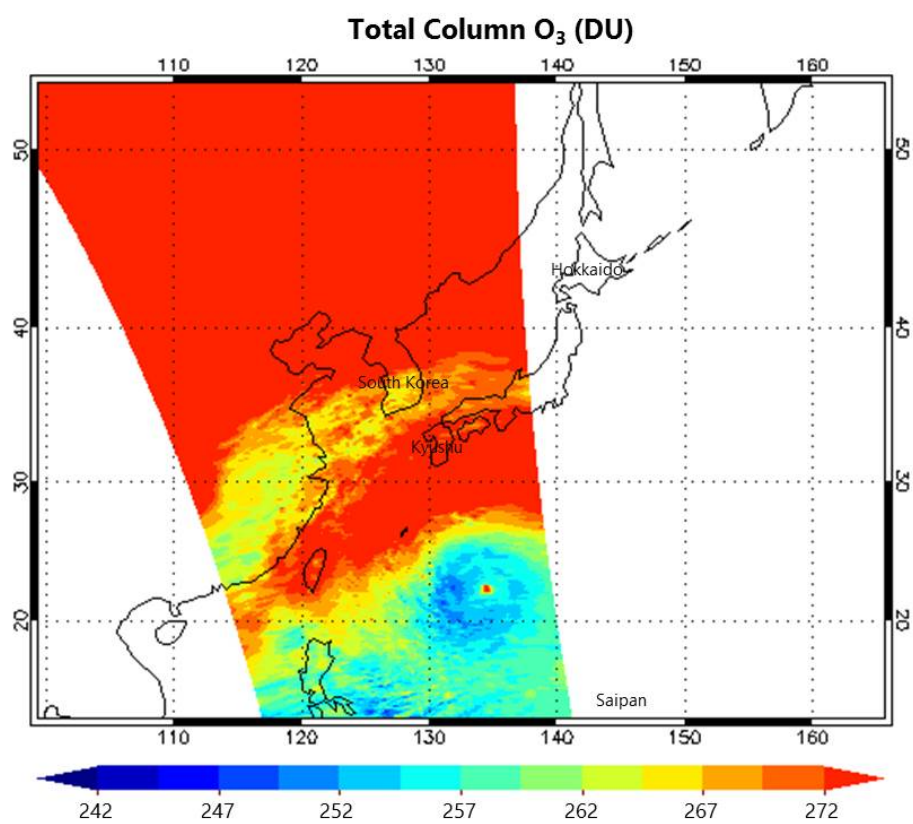


Figure 1. Total column O₃ (in DU) from OMI at 04:05 UTC, 3 September 2005.

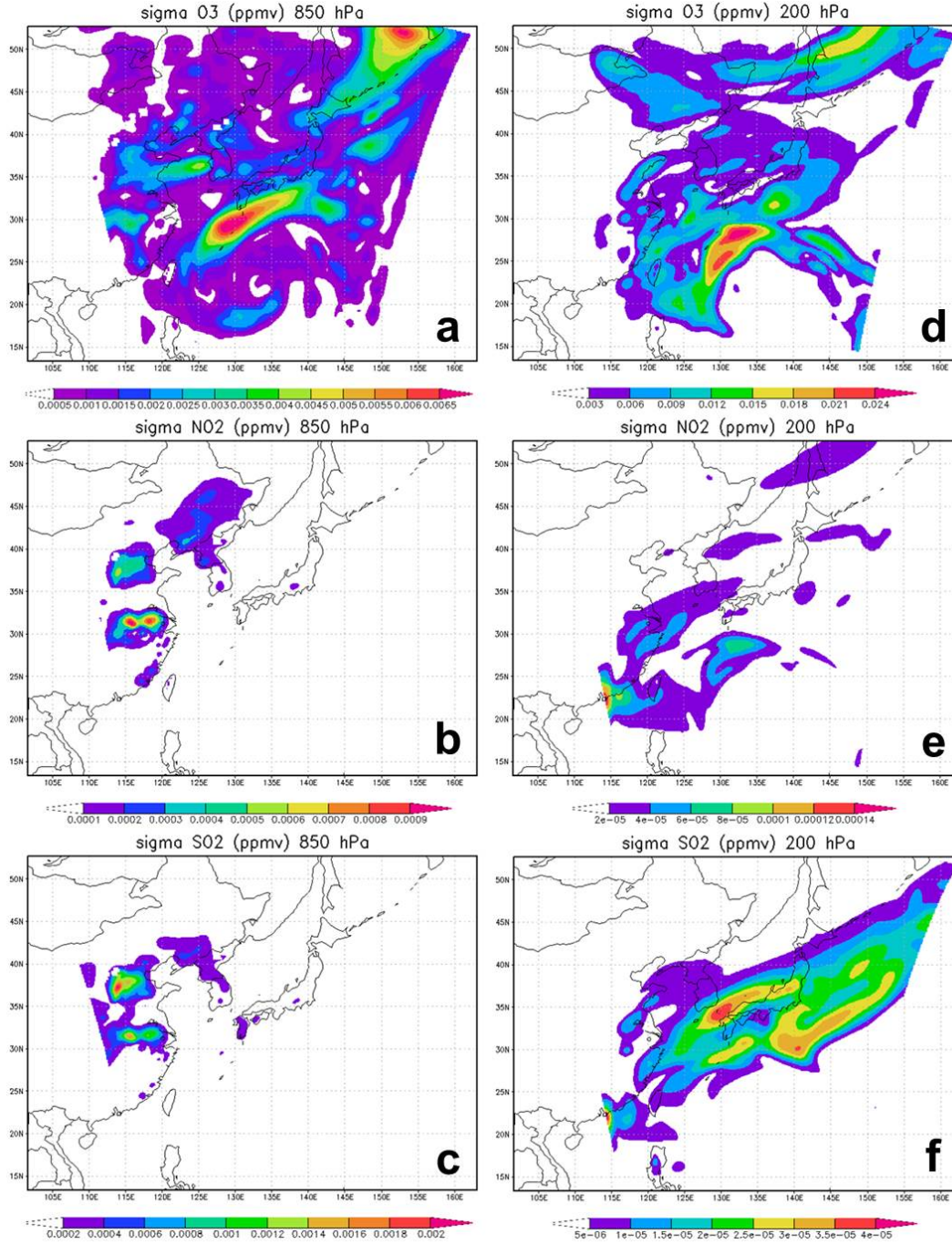


Figure 2. Standard deviation of background error covariance for chemical variables valid on 06:00 UTC, 3 September 2005 at 850 hPa (left panel) for (a) O₃, (b) NO₂ and (c) SO₂, and at 200 hPa (right panel) for (d) O₃, (e) NO₂ and (f) SO₂. Units are ppmv.

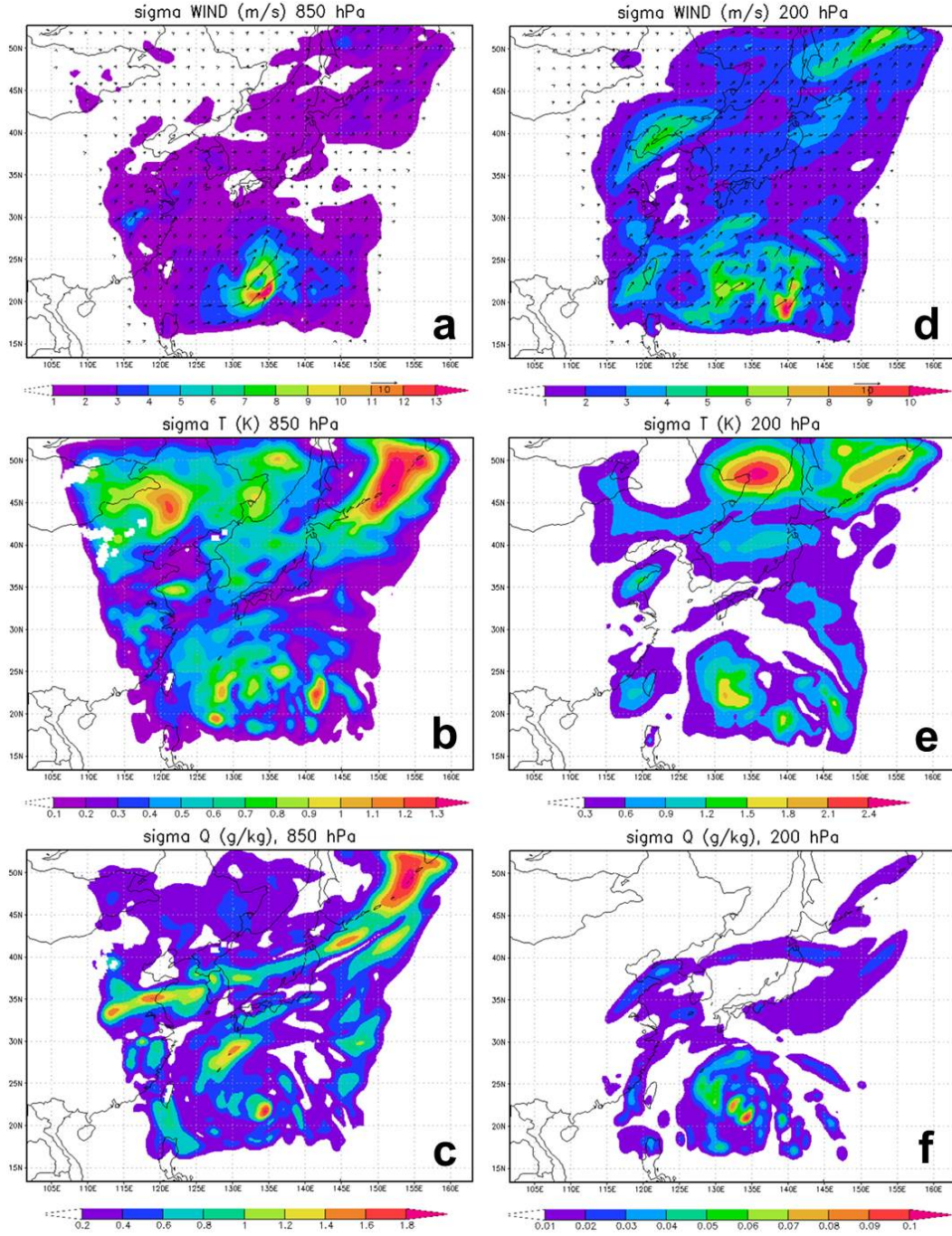


Figure 3. Standard deviation of background error covariance for atmospheric variables valid on 06:00 UTC, 3 September 2005 at 850 hPa (left panel) for (a) wind, (b) temperature and (c) water vapor mixing ratio, and at 200 hPa (right panel) for (d) wind, (e) temperature and (f) water vapor mixing ratio. Units are m s^{-1} for wind, K for temperature and g kg^{-1} for water vapor mixing ratio.

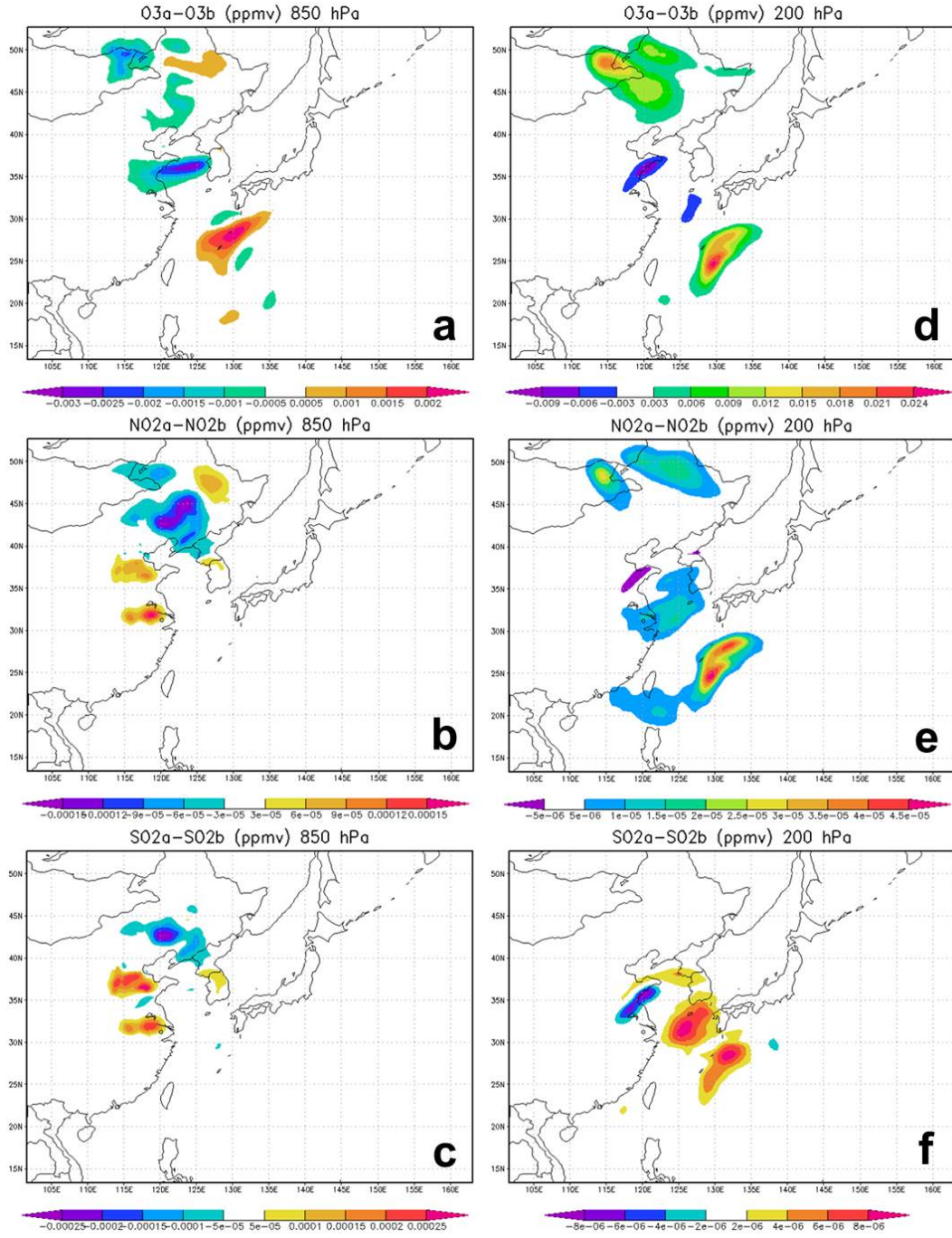


Figure 4. Same as in Fig. 2 except for analysis increment ($x_a - x_b$) of chemical variables in response to total column O_3 . Units are ppmv.

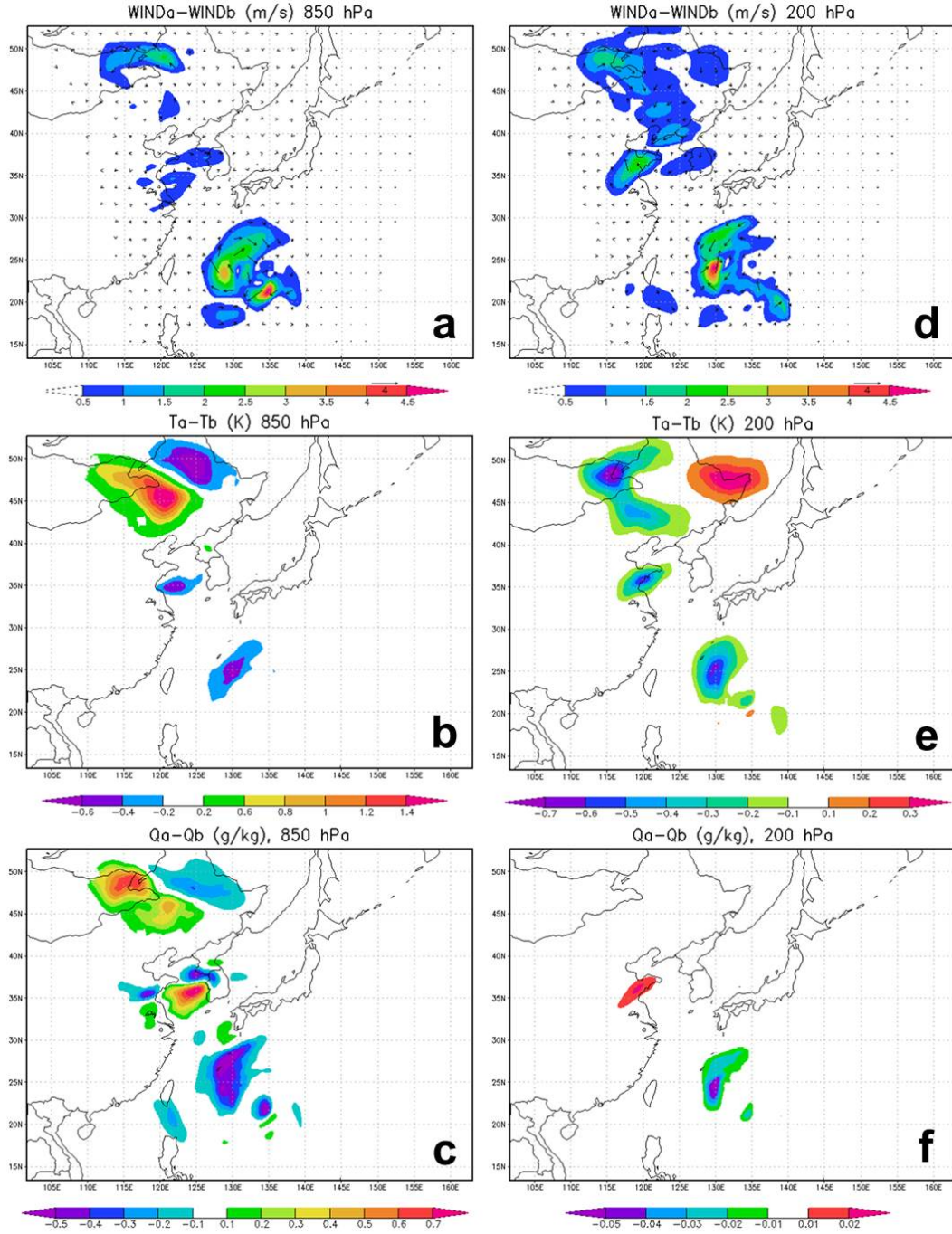


Figure 5. Same as in Fig. 3 except for analysis increment ($x_a - x_b$) of atmospheric variables in response to total column O_3 . Units are $m s^{-1}$ for wind, K for temperature and $g kg^{-1}$ for water vapor mixing ratio.

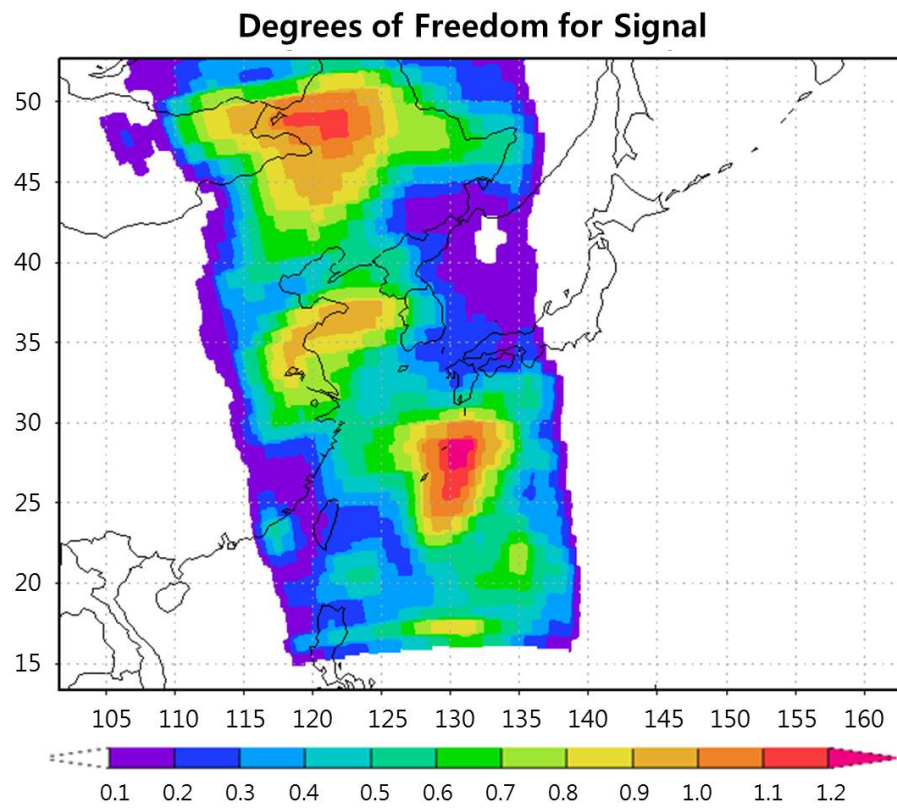


Figure 6. Degrees of ~~Freedom~~freedom for signal (DFS) of assimilated total column O_3 observation valid at 06:00 UTC, 3 September 2005. The units are non-dimensional.