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Improving global chemical simulations of the upper troposphere–lower stratosphere with sequential assimilation of MOZAIC data

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ABSTRACT

The quantity and variety of atmospheric measurements of chemical constituents are strongly increasing, both for in situ and remote-sensing observing systems. This is an incentive to develop data assimilation schemes for atmospheric chemistry models, in the perspective of building realistic three-dimensional time-dependent distributions of observed and chemically related non-observed compounds. Sequential data assimilation experiments within the global Chemistry and Transport Model MOCAGE have been conducted, in the perspective of using the MOZAIC database and meteorological analyses to drive a global simulation of ozone and related tracers, focussing specially on the upper troposphere–lower stratosphere; the chemistry and dynamics in this region of the atmosphere are of great environmental interest, yet they are currently very difficult to model. Assimilation of subsonic flight-level observations of ozone is anticipated to provide a significant contribution to improve numerical simulations. Results obtained with two different sequential data assimilation schemes are presented. The experiments have been set up on an 8-d period in February 1997. The "non-local" data assimilation technique appears preferable to the "local" technique tested, since the benefits of assimilation for the former appear to remain for longer, possibly up to 3–4 d in the model integration after the data assimilation phase.

1. Introduction

MOCAGE (Modèle de Chimie Atmosphérique à Grande Echelle) is the new Météo-France global three-dimensional Chemical Transport Model (CTM), dedicated to the numerical simulation of the interactions between dynamical, physical and chemical processes in the lower stratosphere and in the troposphere (Peuch et al., 1999). MOCAGE is a semi-lagrangian grid-point model with up to four levels of nested domains (two-ways) with increasing horizontal resolutions: the global resolution is generally 2°, while the zoom limited area domains can have resolutions down to 10 km, depending upon the availability of meso-scale meteorological forcings. The model includes 47 hybrid (σ, P) levels from the surface up to 5 hPa. The vertical resolution is about 800 m in the vicinity of the tropopause and in the lower stratosphere. Only the global version of MOCAGE is considered in this study, at a horizontal resolution of 2°. The validation of this new model is currently on-going at Météo-France; however, MOCAGE is developed on the basis of our stratospheric CTM REPROBUS (Lefèvre et al., 1994), sharing the same semi-lagrangian transport scheme which has been thoroughly tested during multi-year simulations (WMO, 1998). A run lasting 7 months of 2000 has been conducted with MOCAGE and REPROBUS: as expected, the two models present similar performances in the stratosphere, while MOCAGE (considering detailed emissions and tropospheric dynamics and chemistry) shows a better agreement with in-situ observations in the upper troposphere–lower stratosphere(UT–LS) (sonde and MOZAIC data) and down to the surface (ground-level stations). MOCAGE

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is currently used for a range of applications in the field of chemical weather forecasts and has been used in real-time during the ESCOMPTE field campaign (see http://medias.obs-mip.fr/escompte).

Data assimilation techniques have been used in numerical weather prediction for several decades (see for instance Bergthorsson and Döös, 1955; Lorenc, 1988). Their application in the field of atmospheric chemistry is more recent (Riishöjgaard, 1996; Stoffelen and Eskes, 1999). Preliminary experiments of sequential data assimilation within the CTM MOCAGE have been conducted, using the MOZAIC database (Marenco et al., 1998) (Measurements of OZone and wAter vapour by aIrbus in-service airCraft; see http://www.aero.obs-mip.fr/mozaic). MOZAIC is a European programme which produces world-wide upper-air ozone and water vapour measurements on a routine basis using five equipped commercial aircraft. It generally takes 3–4 d to have a full coverage of MOZAIC data, which is global, although the Pacific Ocean and Southern Hemisphere receive only limited coverage. Hence, there is a need for data assimilation technique which can have an impact on the model that remains for several days.

An experiment has been set up over 8 d of February 1997 (12 UTC 19 February to 12 UTC 27 February), to test and compare several methods. Ozone photochemistry is parameterized within MOCAGE according to the REPROBUS chemical scheme (Lefèvre et al., 1994), which, in addition to detailed stratospheric chemistry, includes methane oxidation, extending its validity into the upper troposphere. This chemical scheme has recently been used to assess the impact of subsonic aircraft on climate. It comprises 55 transients and species and takes into account heterogeneous chemistry on Polar Stratospheric Clouds, hence affording a realistic ozone hole representation (Lefèvre et al., 1998).

A reference run of MOCAGE (denoted EXP1 hereafter) has been performed over the 8 d, driven by the European Centre for Medium-Range Weather Forecasts (ECMWF) meteorological analyses and without any chemical data assimilation. These ECMWF analyses are based on a global 3D-VAR assimilation technique (Courtier et al., 1998). This global assimilation does not consider ozone, the model resolution is 31 levels in the vertical and T213 in the horizontal (spectral truncation). The 3D initial distributions of chemical tracers have been obtained from a previous spin-up simulation of more than a month and a half, beginning on 1January 1997 from a climatology of the model and using an Equivalent Latitude Initialization (Lary et al., 1995). Two other experiments have been performed over the same 8 days and starting with the same initial chemical tracer distributions, but with data assimilation of MOZAIC ozone measurements over the first 4 d and using two different assimilation techniques. The aim is to check how the corrections brought by the assimilation evolve over the next 4 d, in comparison to the reference, to subsequent MOZAIC ozone observations and to independent ozone radio-soundings available from the World Ozone and Ultraviolet radiation Data Center (http://www.msc-smc.ec.gc.ca/woudc). Thus, this allows a case study of how, and for how long, the assimilation of MOZAIC observations can have an impact upon the model distributions.

Because of the low vertical correlation in the ozone field, it is expected that only the vertical vicinity of flight cruise levels may be affected. However, these levels are generally within the UTLS region, which is of great scientific interest, and yet poorly represented in current state-of-the-art atmospheric chemistry models (Law et al., 2000). Indeed, if the main features of each layer are generally well captured by atmospheric General Circulation Models or CTM, it is not the case for the tropopause region. The UTLS is, however, of importance in the perspective of the assessment of the environmental impact of human activities: it corresponds to the region where subsonic aircraft release exhaust gazes and particles; also, occasional intrusions of stratospheric air into the troposphere bring ozone-rich air masses into the troposphere, and may make a natural contribution to ozone episodes at the surface (Plantevin, 2000).

2. Ozone assimilation experiments

A first assimilation experiment, denoted EXP2, has been conducted using a simple "local" technique: ozone departures computed sequentially between the model and each MOZAIC observation affect the model in the vicinity of flight tracks at each chemical time step (15 min). Several formulations have been tested for the weights given to ozone observation departures, depending upon the geographical distance r between one observation and the model grid-points. For the results presented here, the weight formulation of Cressman (1959) is used, both for the horizontal and the vertical, with observations considered as perfect (i.e. no observation error standard deviation is taken into account):

$$w(r) = \frac{R^2 - r^2}{R^2 + r^2}$$

The computed typical scale of influence of an ozone departure is of the order of $R = 400$ km on the horizontal and of 1.5 km on the vertical.

Very similar results, not presented here, are obtained using a method in which the computation of ozone spatial error correlations has been performed using the MOZAIC database, and in which a representative error term has been added to account for the coarse resolution of the model domain used (2°) compared to the fine resolution of MOZAIC measurements along trajectories.

The second form of data assimilation technique tested in this study is a "non-local" one (referred to as EXP3 in the following). In EXP3, MOZAIC observations are assimilated in a flow-following coordinate system: potential temperature on the vertical, and ozone equivalent latitude on the horizontal. The underlying hypothesis is that the chemical production and destruction of ozone and diabatic processes can be neglected over a period of a few days. We have chosen the ozone equivalent latitude (O3EL) rather than the Potential Vorticity Equivalent Latitude (PVEL), introduced in Lary et al. (1995), because ozone spatial patterns appeared to be readily well captured in the raw model simulation EXP1 (driven by meteorological analyses). The computation of PV and PVEL leads to small numerical structures that can affect the assimilation process, whereas the ozone patterns produced by MOCAGE are fully balanced with the rest of the model. Doing this, what is expected from the assimilation of MOZAIC data is not to affect spatial patterns of ozone in the model, but to modify the ozone gradients according to the observations and to provide a sharper definition of the chemical tropopause and of ozone structures (filaments, local maxima, etc.). In other words, the values of individual isolines of ozone can be modified in the assimilation process, but their horizontal positions are preserved.

A coordinate transformation from model space to assimilation space (and back) is used in this "non-local" assimilation method; it is briefly described here. Firstly, model ozone is projected on potential temperature (θ) isentropes, from 310 to 380 with a 5 K step. Figure 1a provides an example of the ozone field projected onto the 340 K isosurface for the initial date (12 UTC 19 February). Secondly, the ozone equivalent latitude (O3EL) is computed; by straightforward analogy to PVEL (Lary et al., 1995), the equivalent latitude corresponding to an ozone isovalue on a given isentrope is simply defined as the value of the geographical latitude that encompasses a surface area of the same size. In Fig. 1b, the shaded area corresponds to ozone contents greater than 150 ppbv and defines an area which is equivalent to the one between the geographical latitude 35°N and the North Pole; hence the correspondence between the isoline 150 ppbv and the equivalent latitude O3EL = 35°N for the 340 K isentrope. Once the O3EL fields are computed on each of the isentropes, the third step is the mapping of model ozone in the new coordinate system (θ,O3EL): on each isentrope, the model ozone value for a particular O3EL is given by the isovalue that defines that particular equivalent latitude; a resolution of 0.5° is used for O3EL. Given their longitude, latitude and potential temperature, MOZAIC observations are also localized in this (θ,O3EL) coordinate system in order to compute departures and correct the model ozone field. Finally, the computation back from the ozone assimilation field from (θ,O3EL) to the model ozone values on θ isentropes, and then to the original model coordinate system, is performed. As a result, a single observation may have an impact over the whole surface area corresponding to the same O3EL and the same potential temperature: see for example the region shaded in black in Fig. 1a.

This procedure is run every day at 12 UTC over 4 d, using for each daily assimilation all the observations available between 12 UTC and 12 UTC the day before.

Such a scheme is non-centred in the sense that the ozone observations of a 24 h time-slot affect the model at the end of the time-slot. It has been compared with a centred scheme in which the model is affected every day at 12 UTC, but with observations available between 00 UTC and 00 UTC the day after. No significant difference has been found. All observations in the one day time-slot are averaged onto the (θ,O3EL) grid, which provides a set of super-observations, one for each grid-cell. We use a successive-correction method for the assimilation: each super-observation contributes to the analysis within a radius of influence around the grid-point it corresponds to. Weights given to ozone departures are decreasing functions of the distance (in O3EL and θ) between observation and analysis grid-point. These weights correspond both for the horizontal (O3EL) and the vertical (θ) to a 5° polynomial function obtained by a least-squares fitting of experimental error correlations depending on the distance, ΔO3EL and $\Delta\theta$, respectively.

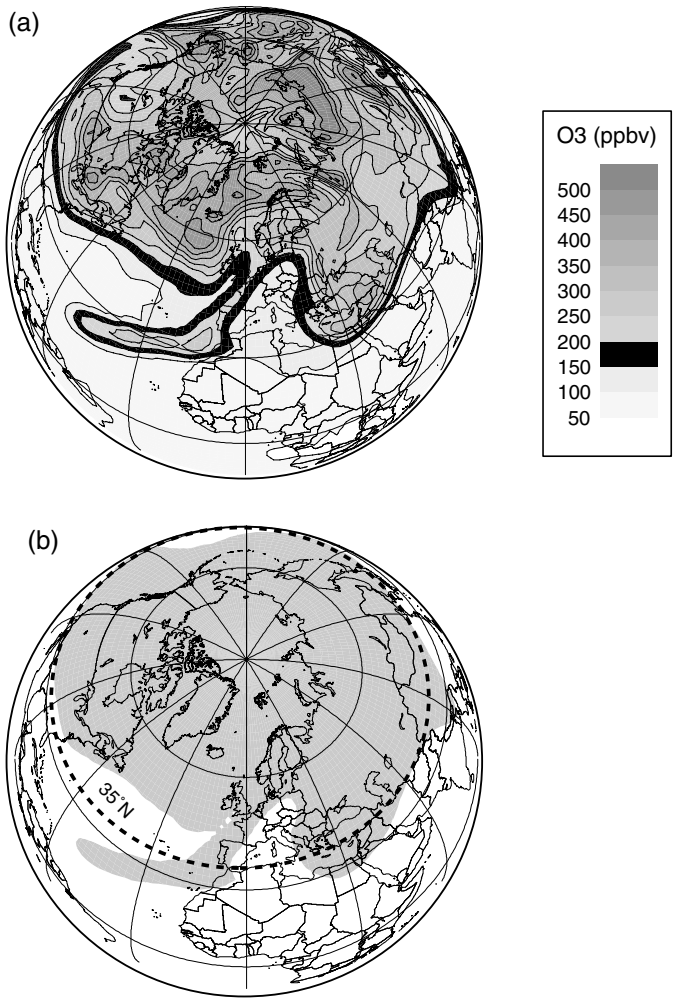

Fig. 1. Principle of the (O3EL, potential temperature) coordinate system. (a) Model ozone isocontours (ppbv) on the 340 K isentrope for 12 UTC 19 February 1997. (b) The shaded surface corresponds to an O3EL greater than 35° and encompasses an area equivalent to that between the geographical latitude circle 35°N and the North Pole. See text for details.

Two consecutive iterations are performed for correcting the model in the (θ,O3EL) coordinate system: the first correction uses a vertical radius of influence $\Delta\theta_1 = 10$ K, the second uses a vertical radius of influence $\Delta\theta_2 = 5$ K. For each correction, the horizontal radius of influence ΔO3EL is computed dynamically according to the local first guess gradient:

$$\Delta\text{O3EL} = \Delta\theta \left(\frac{\Delta\text{O3EL}}{\Delta\theta} \right)_M .$$

3. Results and discussion

Figure 2 presents a zoom on the (80°W to 30°E, 20°S to 60°N) window of the global ozone fields computed for 12 UTC 24 February. This date corresponds to the end of the fifth day of the run, that is one day after the end of assimilation for EXP2 and EXP3. The plot corresponds to the 238 hPa isobar, which is the mean pressure for flight cruise levels. This surface crosses the tropopause, and, as a consequence, shows both stratospheric (principally to the north) and

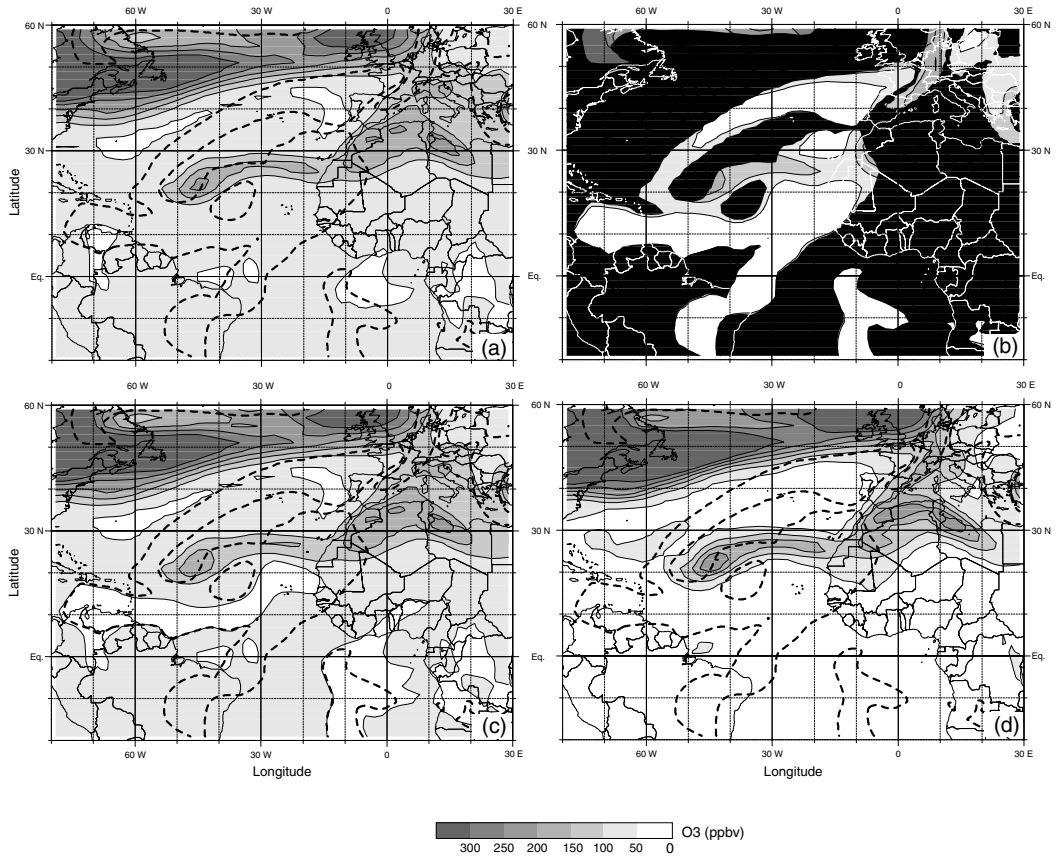

Fig. 2. Model ozone fields (ppbv) at 238 hPa for 12 UTC 24 February 1997, zoomed over the (80°W to 30°E, 20°S to 60°N) window. (a) EXP1, raw model run; (b) "analysis" (see text); (c) EXP2, "local" assimilation experiment; (d) EXP3, "non-local" assimilation experiment. Inside the dashed black lines, the absolute relative difference between the "analysis" and EXP1 is greater than 10%.

tropospheric (to the south) air. A filament of ozone-rich air can be seen in the centre of the domain; although present in the raw model experiment (EXP1, Fig. 2a), this filament is barely marked. We can compare ozone fields at this level from EXP1 (Fig. 2a), EXP2 (Fig. 2c), EXP3 (Fig. 2d) and from an "analysis" (Fig. 2b). We call "analysis" the experiment performed using the local assimilation data of all available MOZAIC flights during the whole 8-d period. This run provides an estimation of the best possible impact of MOZAIC data in the vicinity of the flight tracks. Dashed black lines plotted on the three maps delimit the areas where the relative absolute difference between the "analysis" and EXP1 are higher than 10% (Fig. 2b). During the fifth day (12 UTC 23 February to 12 UTC 24 February), two MOZAIC flights were available in this region and the two corresponding flight tracks appear. These areas within dashed lines are, according to the available observations, the region where the raw model has to be corrected and where we can compare EXP1 with EXP2 and EXP3.

Figures 2a and 2c clearly demonstrate that EXP1 and EXP2 do not differ much and that the assimilation of MOZAIC flights has only brought a transient perturbation to the model, which has almost disappeared after one day: within the dashed lines, the intensity and patterns of ozone only barely differ; the principal difference (of the order of 30 ppbv) is near Caracas, probably generated by the assimilation in EXP2 of the last landing (22 UTC 22 February) and take-off (9 UTC 23 February) of an equipped aircraft in the region. This limited impact of 'local' assimilation is also

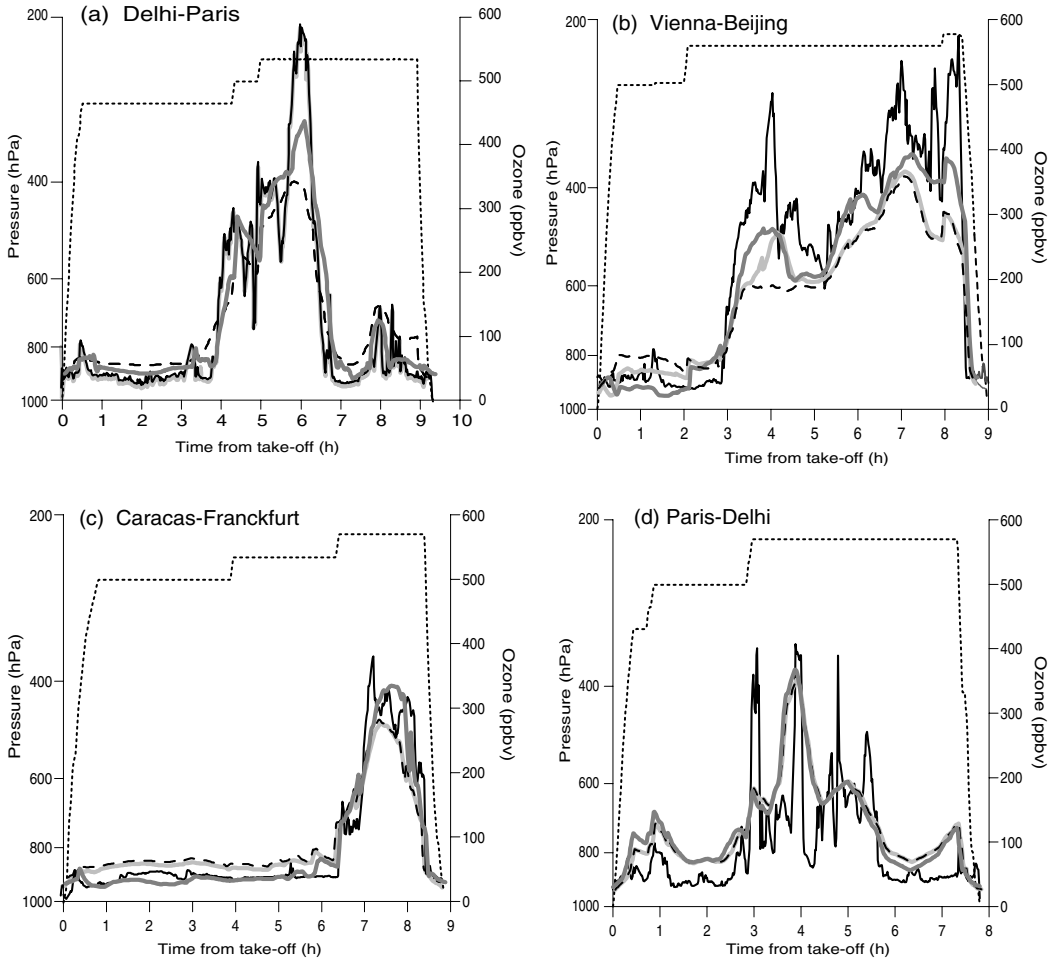

Fig. 3. Model versus four selected individual MOZAIC flights: (a) Delhi–Paris, 2000 UTC 22 February to 0519 UTC 23 February; (b) Vienna–Beijing, 1358 UTC 23 February to 2258 UTC 23 February; (c) Caracas–Frankfurt, 1955 UTC 23 February to 0445 UTC 24 February; (d) Paris–Delhi, 0935 UTC 26 February to 1723 UTC 26 February. Solid black line, MOZAIC observations; dashed black line, EXP1; solid light gray line, EXP2; solid dark gray line, EXP3. The thin dotted black line indicates the flight level (hPa).

demonstrated by Fig. 3c: it is a comparison of model ozone and observations along the Caracas–Frankfurt MOZAIC flight path on the evening of 23 February, that is about 11 h after the landing in Caracas of an assimilated flight. EXP2 (solid light gray lines) presents the same biases as EXP1 (dashed black lines) compared to the in-situ observations (solid black lines): too large ozone contents during the first 6 h when the aircraft flies in the troposphere and too low values when the plane penetrates into the stratosphere, mostly during the last 3 h. A flight-by-flight comparison with all available MOZAIC flights (not presented here) shows that the information brought by the "local" assimilation is only conserved in the model for latitudes above 45°N. At these latitudes, the flight trajectories are principally zonal, and the average large-scale transport helps in keeping information zonally. EXP2 appears to be superior to EXP1 during 1–2 d in these areas. As an example, Fig. 3b shows a Vienna–Beijing flight, one full day after the assimilation of a previous flight in the vicinity. The run EXP2 has reduced tropospheric ozone values and increased stratospheric ones compared to EXP1, and is closer to the observations. However, 3 d after the end of assimilation, no difference

can be made between EXP1 and EXP2 along the Paris–Delhi flight (Fig. 3d): the information brought by the "local" assimilation is lost.

By sharp contrast, EXP3 differs from EXP1. Figure 3a shows that during the assimilation period, as expected, the "local" assimilation method brings the model close to assimilated observations, whereas the "non-local" technique only corrects large-scale error patterns. However, comparing Figs. 2b and 2d, it appears that one day after the end of assimilation, EXP3 is remarkably similar to the "analysis" in areas inside the dashed lines. EXP3 presents reduced tropospheric ozone values compared to EXP1, and captures the ozone filament at 25°N well. The improvement can be quantified on the Caracas–Frankfurt flight (Fig. 3c): the tropospheric ozone levels are perfectly seen, and the region of the filament, as well as the stratospheric part of the flight, are clearly improved. Figures 3b, 3c and 3d present a first illustration of the benefits of "non-local" assimilation over "local" assimilation, the impact of which appears to last longer. More than 3 d after the end of assimilation, EXP3 differs from EXP1. This is specially interesting since it takes 3–4 d to have a complete coverage with MOZAIC flights, with at least one flight for each destination. The "non-local" assimilation method can affect the model over a period of time of about the same duration and makes it possible to consider preparing a long simulation of the model, which will take full advantage of the assimilation.

This result can be confirmed on the basis of a statistical analysis of the last 4 d of the simulation (i.e. the period without assimilation). In Fig. 4a, MOCAGE ozone values are displayed as a function of MOZAIC ozone values for EXP1, EXP2 and EXP3. It can be seen that EXP2 and EXP3 are closer to ideality than the experiment without assimilation (EXP1), although the model biases behave in a similar way: the model tends to overestimate low ozone contents and to underestimate high ozone values. For high values, encountered mainly when the flight enter air masses in the stratosphere or of stratospheric origin, the relatively coarse resolution of the model (2° horizontally and 800 m vertically) is probably the main reason explaining why the maximum values are underestimated. For low ozone values, which correspond to the samplings during landing, take-off and transit within the troposphere, the slight overestimation could be related to the fact that the model is unable to represent the ozone destruction by nitrogen oxides in urban or heavily industrialized areas where international airports generally lie. Globally, on this 4 d period, the "local" data assimilation scheme (solid light gray line) is not as successful as the "non-local" one (solid dark gray line) in reducing the model biases compared to subsequent MOZAIC measurements.

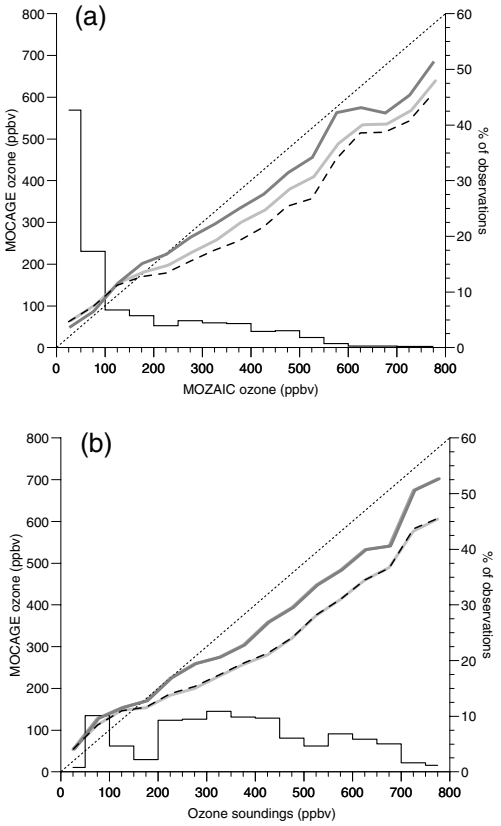

Fig. 4. Mean bias between model ozone and observations for the four last days of the numerical experiments (12 UTC 24 February to 12 UTC 27 February). (a) observations = all available MOZAIC data of the period; (b) observations = all available ozone radio-soundings (taken only between 280 and 180 hPa) of the period. Dashed black line, EXP1; solid light gray line, EXP2; solid dark gray line, EXP3; thin black line, percentage of observations in each 50 ppbv bins. The thin dotted black line indicates the ideality.

The benefits to the three-dimensional (3D) model ozone distributions due to assimilation have also been investigated using ozone radio-soundings, which were obtained from the WOUDC. These in-situ ozone data provide an independent set of observations covering the globe, including regions MOZAIC flights never document. The general diagnostic of the model error over the last 4 d is the same: underestimation of

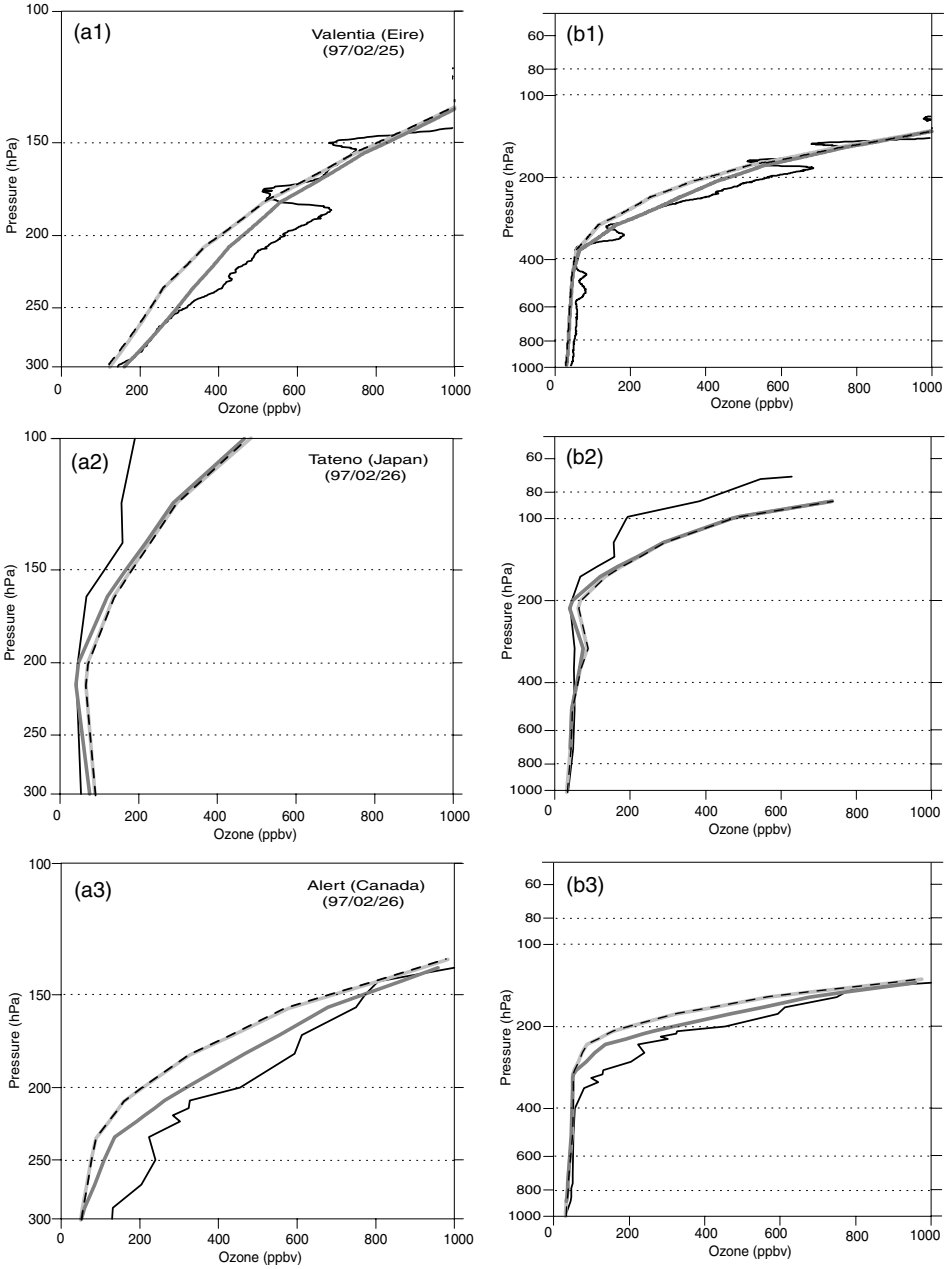

Fig. 5. Observed and modeled ozone (ppbv) for: (a) profiles between 300 hPa and 100 hPa; (b) the corresponding entire vertical profiles. (a1) and (b1) Valentia (Eire; 52°N, 10.3°W), 0845 UTC 25 February; (a2) and (b2) Tateno (Japan; 36°N, 140°E), 0600 UTC 26 February; (a3) and (b3) Alert (Canada; 82.5°N, 62.3°W), 1400 UTC 26 February. Solid black line, ozone radio-sounding; dashed black line, EXP1; solid light gray line, EXP2; solid dark gray line, EXP3. The thin dotted black line indicates the altitude (hPa).

high ozone values and overestimation of low values. However, against those independent data, only EXP3 appears to bring an improvement compared to the raw model situation (EXP1), as shown in Fig. 4b. The "non-local" assimilation not only provides a longer conservation of the increments, but also has an interesting impact over the whole global distributions, even in the parts of the Southern Hemisphere, which is nevertheless little observed by the MOZAIC flights.

Verifications against individual ozone soundings confirm this statistical result. Figures 5a1, 5a2 and 5a3 present three ozone profiles between 100 and 300 hPa (MOZAIC flights levels), comparing EXP1, EXP2 and EXP3 profiles. The corresponding entire ozone profiles are also shown in Figs. 5b1, 5b2 and 5b3. EXP3 compares more favorably with observations than the other experiments. Again, this result remains valid up to 3 or 4 d ahead of the assimilation periods.

After these results, another assimilation technique has been verified, combining the non-local correction scheme with the local one. It seems that the local assimilation, when added to the non-local one, introduces small-scale patterns which destroy the large scale ozone consistency which is usually well captured by the model. Then, the next non-local assimilation is adversely affected by these small-scale patterns. Results, not reported here, show that the combination of these two techniques is worse than the non-local one.

4. Conclusions and perspectives

Two different types of data assimilation schemes have been tested for assimilating MOZAIC ozone data in the global 3D CTM MOCAGE: analysis correction techniques are used in both types of schemes. However, the first type is a local correction method, whereas the second type is based on a change of coordinates to potential temperature and ozone equivalent latitude. The second type of scheme is essentially non-local and exploits the quasi-conservative property of ozone in the new coordinate system.

Different tunings have been tested inside each type of methods, and they have shown little impact on the results. The non-local techniques appear to be superior for assimilating MOZAIC data with respect to all the verification diagnostics, which have been developed: verification against the MOZAIC data themselves, against independent ozone radio-soundings (not used in the assimilation). The more promising result is that the corrections to the model ozone introduced by the MOZAIC flights through the non-local assimilation scheme are kept for 3–4 d after the actual use of the MOZAIC data. This "conservation time" is much shorter for a local assimilation technique.

Hence, a long MOCAGE simulation including a non-local data assimilation of MOZAIC observations is in preparation. This will provide an unprecedented 4D dataset for ozone with a realistic UTLS. Moreover, the model horizontal resolution aspects will also be addressed because it is known that some small-scale structures are missed by the $2^\circ \times 2^\circ$ resolution model and captured when increasing the horizontal resolution. Consequently, it is envisaged to check what are the benefits of a non-local assimilation with resolutions of $1^\circ \times 1^\circ$ and $0.25^\circ \times 0.25^\circ$ over limited area domains available within MOCAGE.

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