

Chemical Transport Modelling

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1. Introduction

Nowadays, a large community of modellers use chemical transport models (CTMs) routinely to investigate the distribution and evolution of tracers in the atmosphere. Most CTMs use an ‘off-line’ approach, taking winds and temperatures from general circulation models (GCMs) or from meteorological analyses. The advantage of using analyses is that the CTM simulations are then linked to real meteorology and the results are directly comparable to observations. Reanalyses extend this advantage into the past, allowing us to perform long-term simulations that provide valuable information on the temporal evolution of the atmospheric composition and help understand the present and predict the future. CTMs therefore rely on the quality of the (re)analyses to obtain accurate tracers distributions. And, in its turn, this reliance makes CTMs be a powerful tool for the evaluation of the (re)analyses themselves.

In this paper we discuss some of the main issues investigated by off-line CTMs and the requirements that these studies have for future (re)analyses. We also discuss tests performed by CTMs in order to evaluate the quality of the (re)analyses, to show in particular the recent improvements achieved in terms of stratospheric transport when the new ECMWF reanalysis winds are used for long-term simulations.

2. Past and present CTMs experiences with (re)analyses

2.1. Long-term ozone loss and stratospheric transport

The ozone loss detected over the past 25 years has important implications, given the strong interactions between stratospheric ozone, UV radiation, circulation, tropospheric chemistry, human and natural activities. For these multiannual stratospheric studies, an accurate representation of the Brewer-Dobson (B-D) circulation is essential so that CTMs achieve realistic tracer distributions. However, in recent years several studies based on long-term CTM simulations have shown that winds from analyses can produce too strong a stratospheric circulation. ERA-40 [Uppala et al., 2005] and ECMWF operational winds have been reported to produce a much too young age-of-air [e.g. van Noije et al., 2004; Meijer et al., 2004; Scheele et al., 2005; Chipperfield, 2006]. U.K. Met Office (UKMO) winds [Swinbank and O'Neill, 1994] have produced ages even younger than ERA-40 [Chipperfield, 2006]. The underestimation of the age of air implies that the models overestimate the B-D circulation, which clearly affects their ability to reproduce observed tracer distributions.

To correct for this overestimation of transport, several strategies have been adopted by CTM modellers. These include the use of forecasts instead of analyses [i.e. Scheele et al., 2005], the use of 3-hourly instead of 6-hourly winds [i.e. Bregman et al., 2006; Berthet et al., 2005], or the use of different vertical advection and isentropic coordinates [Chipperfield, 2006]. However, for a number of reasons, it is much more desirable that

the analyses themselves are good enough to produce a realistic stratospheric circulation when used directly into CTMs.

In order to solve this overestimation of the B-D circulation, and other detected deficiencies in ERA-40, the ECMWF is preparing a new reanalysis (ERA-Interim) that will succeed ERA-40 for the period 1989-onwards. Results obtained with an experimental run of ERA-Interim (EXP471) using the TOMCAT/SLIMCAT chemistry transport model clearly show the significant improvements achieved by using the new winds for multiannual stratospheric simulations [Monge-Sanz et al., 2006]. Our age-of-air results are now much closer to the observational values [Andrews et al., 2001; Ray et al., 1999] than with ERA-40, and our trajectory calculations show a significant reduction in the excess of mixing in the lower stratosphere (LS) region presented by the new winds with respect to previous analysed winds (e.g. ERA-40, UKMO or GEOS-4 winds). Nevertheless, in spite of the important improvements, a comparison between a TOMCAT run (sigma-pressure coordinate system and divergence for the vertical motion) and a SLIMCAT run (hybrid sigma-isentropic coordinate system and vertical motion from heating rates) indicates that the latter still corrects for some excess of transport in the analysed winds, although to a much lesser extent than with ERA-40 winds [Chipperfield, 2006].

2.2. Polar ozone loss and polar temperatures

One of the key steps in the destruction of ozone over polar regions involves the heterogeneous chemistry that takes place on polar stratospheric clouds (PSCs). These clouds can form only at the low temperatures found inside the polar vortex in winter and spring. Therefore, to correctly simulate the polar ozone loss, CTMs need accurate temperatures over the poles to correctly calculate the areas covered by PSCs. Current (re)analyses however exhibit problems in the temperature field that make the calculation of realistic PSC areas impossible, seriously affecting the polar ozone loss studies.

ERA-40 has been reported to present a cold bias in the upper stratosphere, as well as unrealistic oscillations in the vertical structure of temperature [Randel et al., 2004; Simmons et al., 2005], especially large over the South Pole. Also studies comparing temperatures from different reanalyses against temperatures from radiosondes [e.g. Knudsen, 1996, 2003] reveal the existence of an important cold bias over the Arctic for ERA-40, while NCEP/NCAR is too warm compared to radiosondes in this region [Knudsen, personal communication]. On the other hand, NCEP/NCAR reanalysis was designed for tropospheric studies and is not recommended for polar investigations due to temperature biases in the LS [Manney et al., 2005].

Over the Arctic, the reanalyses from the Free University of Berlin (FUB), from 1965 until at least part of the 1990s, present the best agreement with radiosonde temperatures. It might be surprising that also for the satellite era the FUB analyses are better than the automatised analyses (ERA-40 and NCAR), however one feasible explanation is that when radiosonde data are sparse, the analysis rely on modelled temperatures and satellite derived temperatures. If this goes on for a long time, radiosonde temperatures could be flagged as erroneous and not used in the assimilation [Knudsen, personal communication]. Also the weighting functions for the assimilated satellite data can lead to erroneous vertical distribution of temperatures. It is therefore desirable that future (re)analyses include as much radiosonde data as possible to get a more realistic temperature structure over the poles, which will result in more realistic PSC area calculations.

2.3. Stratosphere to troposphere exchange

Stratospheric transport also affects the budgets of tracers in the troposphere as it controls the exchange between stratosphere and troposphere (STE). In this section we briefly discuss ozone exchange flux calculations from van Noije et al. [2006]. The ozone fluxes are calculated with the linearised ozone model Linoz [Mc Linden et al., 2000] implemented in the KNMI chemical transport model as described in van

Noije et al. [2004]. Calculations have been made with ERA-40 reanalyses for the whole 1957-2002 period, and also with the ECMWF 4D-Var operational analyses for 2000-2005.

The time series of ozone monthly STE fluxes obtained with ERA-40 presents many temporal inhomogeneities. And for the overlapping period, ERA-40 systematically overestimates the flux with respect to operational analyses. STE fluxes are reduced when forecasts instead of (re)analyses are used. Especially for ERA-40 there is a very clear correlation between the reduction in the annual total ozone flux and the increase in the forecast range. This relationship is not so clear for operational analyses, however.

Also in van Noije et al. [2006] an interesting comparison is performed between ERA-40 and a short (Jan-Mar 1973) experimental run of ERA-40 in which no satellite radiances were assimilated. For this experimental run the STE of ozone is lower over the Northern Hemisphere than for the normal ERA-40 run.

Thus, it appears that, for ERA-40, STE fluxes show sensitivity to both forecast range and satellite observations. The improvements in the data assimilation system (4D-Var operational analysis v. ERA-40) clearly improve STE calculations.

2.4. Tropical tropopause layer

The tropical tropopause layer is of particular interest as processes in this region play a key role in the dehydration of water vapour while entering the stratosphere, and are then responsible for the water vapour budgets in the atmosphere.

The work by Krüger et al. [2006] is a representative example of the use of (re)analyses for TTL studies. In this work, they study the variability of water vapour in the TTL by running a trajectory model with ERA-40 and ECMWF operational (re)analyses. The vertical motion is derived from heating rates from an off-line radiation code [Morcrette et al., 1998], to avoid the too noisy analysed vertical velocity field. This Lagrangian model was run backwards for 3 months (Feb-Dec), for 40 different years (1962-2002) for ERA-40, and for 5 years (2000-2005) for operational winds. The minimum temperature (T_{\min}) for each trajectory (5580 trajectories on a $2^\circ \times 2^\circ$ grid) is stored. This method reveals information on the dehydration points of stratospheric water vapour. An intercomparison for the winter 2001/2002 shows that temperatures are lower in operations than in ERA-40, probably due to a cold bias in the tropical stratosphere. For the winter 2005/2006, the current ECMWF system (T799/L91) and the previous one (T511/L60) overlapped and it was possible to compare the T_{\min} distribution obtained with both analyses. This comparison showed that the cold bias in the TTL has been reduced in the present ECMWF system.

One of the main problems in ERA-40 is the excessively dry lower and middle stratosphere [Uppala et al., 2005]; for future (re)analyses, improving the vertical advection (reduction of noise in vertical winds) and improving the temperature structure (reduction of biases) will improve the representation of the water vapour transport across the tropical tropopause.

3. Requirements for future (re)analyses

3.1. Improvements still needed

Results from the studies included in this paper have shown that the new ECMWF system, both for operational analyses and for the ERA-Interim reanalyses, presents important improvements with respect to previous operational systems and to ERA-40 reanalysis. It is, however, still important to stress what essential points need to be further developed in new (re)analyses:

- a) Keep improving the long-term stratospheric Brewer-Dobson circulation.
- b) Improvement in polar temperatures, including more radiosonde data in the reanalyses.

- c) (Re)Analysis constraint to reduce the large uncertainties in the STE fluxes.
- d) Reduction of noise in the vertical winds.
- e) Temperatures in the TTL region should be improved as the operational ones.
- f) Archiving analysed winds every 3 hours instead of every 6 hours. Nevertheless, we would recommend prioritise the quality of the analysis over the increase in the temporal resolution of the archived fields.

3.2. Data availability

It is desirable that physics and dynamics within the CTM are as similar as possible (ideally identical) to their analogous in the meteorological model used to produce the (re)analyses. For this reason, parameters such as heating rates and eta-dot vertical velocities need to be available from the archived analyses for consistency of the horizontal and vertical motion. Besides, CTMs include parameterisations for convection and other physical processes that require certain parameters (mass fluxes, updraft, entrainment/detrainment rates...) to be regularly archived to make the parameterisations in the CTMs as consistent as possible with the equivalent parameterisations in the meteorological model.

In the ECMWF case, these parameters are available for ERA-40 but, for archive space reasons, are not stored operationally. However, reanalyses like ERA-40 have a limited length in time, which poses an important problem for trend studies, in which the most recent meteorological data need to be included without changes in the version of the analyses system.

3.3. Trend studies

Trend studies need to be version independent, as changes in the version of the analyses model mask at least part of the detectable trend evolution.

One example is the recovery of stratospheric ozone over midlatitudes expected due to the detected decrease in atmospheric chlorine content after the Montreal Protocol agreement entered in force in 1989. For this study, the SLIMCAT CTM was run with ERA-40 winds and temperatures until 2002 when it had to switch to ECMWF operational analyses. The problem is that the big difference in the temperature field between ERA-40 and operations completely masks the expected effect of the chlorine reduction.

The CTM community strongly recommends some kind of maintenance to keep reanalyses updated. For instance, updating the reanalyses twice a year to include the most recent dates with the original model used to create the main part of the reanalyses would avoid this kind of problem.

4. Summary

CTMs are currently dealing with key atmospheric issues, relying for these investigations on the quality of the (re)analyses used to prescribe the meteorology. It is important to note that the relationship between CTMs and reanalyses is bidirectional: not only do CTMs need accurate reanalyses, but also CTMs can efficiently help to improve reanalyses by spotting problematic aspects, as the meridional stratospheric circulation; here the longer stratospheric experience of CTMs is a valuable advantage for meteorological analysis systems that have recently extended their vertical range into the stratosphere. The number of CTM modellers that need reanalyses is continuously growing, archiving the fields they require and making these data publicly available will positively increase the use of NWP products.

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