Estimation of mixing in the troposphere from
Lagrangian trace gas reconstructions during
long-range pollution plume transport

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Abstract. The dispersion and mixing of pollutant plumes during long-range transport across the North Atlantic is studied using ensembles of dif-

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fusive backward trajectories in order to estimate turbulent diffusivity coefficients in the free troposphere. Values of the order of $0.5 - 1 \text{m}^2 \text{s}^{-1}$ and $1 \times 10^4 \text{m}^2 \text{s}^{-1}$ for the vertical and horizontal diffusivity coefficients $D_v$ and $D_h$ respectively have been derived. Uncertainties related to the method are discussed and results compared with previous estimates of mixing rates in the atmosphere. These diffusivity estimates also yield an estimate of the vertical:horizontal aspect ratio of tracer structures in the troposphere. The representation of sub-grid mixing in the global Eulerian Chemical Transport Model MOCAGE is also assessed, and optimal time and space grid resolutions required to simulate the long-range transport of pollutants are investigated on inter-continental scales, suggesting the need for vertical/horizontal resolutions of the order of 500m/40 km for this case study. This work is the basis of a detailed study including chemical transformations along ensembles of diffusive backward trajectories [Real et al., 2008].
1. Introduction

Simulations of global atmospheric composition designed to investigate climate change and regional air quality issues rely on numerical models of transport and chemistry. This requires accurate simulation of the long-range transport of pollutants from source to receptor regions. Observations have shown that, downwind from emission regions, where pollutants are uplifted into the free troposphere by convection or frontal systems, differential advection, due to stratification, reduces the scale of tracer structures leading to the formation of narrow sheet-like layers downwind from continents occupying a large part of the volume of the troposphere [Newell et al., 1999]. The variability of tropospheric mixing on small scales plays an important role in the distribution of such layers [Colette and Ancellet, 2006]. The ability of global Eulerian models to simulate the inter-continental transport of pollutant layers is governed largely by the spatial resolution of the simulations. Current computational constraints generally limit global chemical transport models (CTMs) to spatial scales of 1° or greater, with trace gas concentrations assumed constant in model grid boxes. Mixing tends to be overestimated in such coarse resolution models with effective horizontal diffusivities, related to spurious numerical diffusion, leading to the smearing out of small-scale features [Tan et al., 1998]. In addition, chemical rates, calculated from such mean grid values ignore any sub-grid scale variability and correlations between species which can result in systematic errors in CTM trace gas budgets [Pyle and Zavody, 1990; Wu et al., 2007] which will be aggravated in case of nonlinear reactions. Such systematic errors have been reported for ozone [Wild and Prather, 2006], NO$_x$ [Cook et al., 2007], OH [Crowther et al., 2002] and ClO$_x$ [Edouard et al., 1996; Tan et al., 1998].
Other critical issues include the representation of emissions [Sillman et al., 1990; Esler et al., 2004] and sub-grid scale processes such as convection. Model estimations of O$_3$ are clearly sensitive to such processes and the resolution of the simulations [Wild and Prather, 2006] but global models have yet to be tested at resolutions where they reach the limits dictated by small-scale turbulence in the atmosphere.

Small-scale structures can be explicitly represented in a Lagrangian framework, which separates advection by the flow from the effects of chemistry and mixing [Methven et al., 2003]. Trajectory calculations are able to simulate more accurately the structure that results as air masses from different origins are brought into proximity although individual trajectories may be subject to positional errors of +/- 20% [Stohl et al., 2004, 2005]. Pure Lagrangian techniques neglect mixing yielding too large gradients [Legras et al., 2003; Methven et al., 2003], and do not represent chemical reactions leading to changes in concentrations within air parcels. In real flows, the reduction in scale of filamentary structures combined with irreversible small-scale turbulence are efficient mixing processes represented by a turbulent diffusivity tensor. In general, the horizontal component of the diffusivity, $D_h$, is several orders of magnitude larger than the vertical diffusivity, $D_v$ due to quasi isentropic advection associated with strong stratification. This problem has been studied extensively in the stratosphere [Hall and Waugh, 1997; Pisso and Legras, 2008]. Previous estimates in the free troposphere focused on horizontal diffusivities of power plant or aircraft plumes [Sillman et al., 1990; Schumann et al., 1995].

This paper forms the first part of a study designed to investigate the dynamical and chemical processes acting during long-range transport of a pollutant plume in the free troposphere. Here, we focus on the quantification of plume dispersion during long-range
transport and its relation to other mixing estimates including the “mixing” inherent in global models. We focus on the long-range transport of a forest fire plume during summer 2004 when fires were very active over Alaska and large amounts of trace gases such as CO were emitted (e.g. Pfister et al. [2005]) providing an ideal tracer for investigating plume transport and dilution in the troposphere. Multiple aircraft samplings of the forest fire plume during transport from Alaska to western Europe are used to constrain estimations of vertical and horizontal diffusivities calculated using a stochastic Lagrangian reconstruction method previously applied in the stratosphere [Pisso and Legras, 2008]. These results, which allow quantification of dispersion rates of pollutant plumes in the troposphere, are compared to diffusion rates in global 3D models, which are, as noted above, related to numerical diffusion at current resolutions. Results are also compared to other parameters used to describe plume dilution such as mixing rates estimated assuming an exponential decay to background concentrations (e.g. Arnold et al. [2007] and Real et al. [2007]).

In a companion paper [Real et al., 2008], the results presented here are used to perform high resolution photochemical calculations along ensemble multiple trajectories originating every 15-30s along the flight tracks. The objectives were to examine the impact of multiple air mass origins on different parts of the the plume samplings for a well defined case of long-range transport, and to examine the impact of transport versus photochemistry on concentration distributions across the plume. Another aim was to evaluate the importance of local errors in net $O_3$ production in global models related to chemical non-linearities and spatial resolution.

The data and methods used in this work are described in section 2. Estimates of vertical diffusivity from stochastic reconstructions in the troposphere discussing possible sources
of error are presented in section 3. The representation of large-scale advective processes affecting the quality of such reconstructions, in particular tracer gradient formation are discussed in section 4. The results are discussed and compared to previous mixing estimates in section 5. The implications of our findings for global Eulerian chemical models are discussed in section 6 and conclusions are presented in section 7.

2. Data and Methods

2.1. The Lagrangian 2K4 campaign

During ICARTT (International Consortium for Atmospheric Research on Transport and Transformation), in summer 2004, a series field experiments were conducted to study the processes influencing long-range transport of trace gases and aerosols over the North Atlantic from North America to Europe (see Fehsenfeld [2006] for further details about flights and aircraft instrumentation). A Lagrangian pollutant experiment (International Global Atmospheric Chemistry (IGAC) Lagrangian 2K4) over inter-continental scales was performed with as many as 6 successful multiple samplings of the same air masses during transport across the North Atlantic [Methven et al., 2006].

One of these cases involved the multiple sampling of a forest fire plume transported from Alaska to the west coast of Europe. This plume, clearly visible in MOPITT CO data stretching over the North Atlantic [Bousserez et al., 2007] may have been lofted into the free troposphere by convection triggered by the passage of a low pressure system following a dry and unusually warm period [Damoah et al., 2006]. The Lagrangian samplings by several aircraft have been analyzed in detail by Methven et al. [2006] (case 5) and Real et al. [2007]. Using backward and forward trajectory analysis and hydrocarbon fingerprint matching, Methven et al. [2006] identified Lagrangian matches between flight segments.
on the 18 July during a DC8 flight over Newfoundland and a DLR Falcon flight on 23 July over the English Channel, five days later. Further trajectory analysis by Real et al. [2007] also showed a match between the DC8 plume match on the 18 July and a DLR Falcon sampling on 22 July off the coast of Spain and southwest France. Real et al. [2007] also examined the processes responsible for the evolution of $O_3$ in the plume using a photochemical box model initialized with upwind observations. Whilst, photochemical production of $O_3$ due to PAN decomposition was important when the plume descended toward Europe, mixing with other air masses, based on the decrease of CO in the plume, was also shown to be an important factor governing trace gas evolution. These results are compared with this study in section 5.

2.2. Lagrangian trajectory calculations

2.2.1. Diffusive backward trajectories

Atmospheric flows exhibit strongly stratified dynamics outside convective regions. Hence, the main contribution to mixing comes from small-scale turbulent motion. This motivates the representation of small-scale turbulence in Lagrangian calculations as stochastic perturbations on the velocity field as a way of taking into account perturbations occurring at sub-grid scales. It is assumed that the overall effect of turbulence in a spatial scale smaller than the grid of the advecting fields has time scales much smaller than the time step of advection, and hence can be approximated as a diffusive process. Due to the typical aspect ratio of atmospheric structures, this stochastic perturbation can be added either to the vertical or horizontal advecting winds.

Reverse integrations of ensembles of trajectories initialized along the Lagrangian match segments of the flight tracks have been performed with TRACZILLA [Legras et al.,
2003, 2005; Pisso and Legras, 2008]. This is a modified version of the trajectory model FLEXPART [Stohl et al., 2005] which includes vertical or horizontal independent stochastic perturbations in the velocity field. The model was run using winds from the European Center for Medium Range Weather Forecasts (ECMWF) operational analyses at 1° horizontal resolution, and on 60 hybrid levels with 3-hour resolution obtained by combining analyses available every 6 hours with short time forecasts at intermediate times (3 hour resolution). One thousand parcels were initialized every second along the flight segments. CO concentrations from the global model MOCAGE (see next subsection) were mapped onto the backward endpoints of the trajectories, 4 to 9 days prior to the plume samplings. The stochastic perturbations associated with the turbulent diffusion in the vertical, $D_v$, or horizontal, $D_h$, applied to parcel positions, were varied in order to obtain the best agreement between stochastic reconstructions and the measured CO data along the flight tracks.

These diffusive reconstructions, based on stochastic ensembles of backward trajectories, use dynamical information contained in the time series of advecting winds. This “zooming” translates this temporal information into increased spatial resolution of tracer fields from three dimensional CTMs leading to improved agreement with fine scale features observed in the in-situ measurements. Moreover, the resulting averaged vertical velocity helps to reduce uncertainties associated with the vertical velocity ($\omega$) field.

### 2.3. MOCAGE

Global CO fields from the MOCAGE (MOdel of atmospheric Chemistry At larGe Scale) 3-D CTM were used to initialize the multiple trajectory calculations. MOCAGE uses a semi-Lagrangian advection scheme [Josse et al., 2004] to transport chemical constituents
or tracers. The horizontal grid resolution used in this simulation is 2 x 2 degrees. The model includes 47 hybrids ($\sigma - p$) levels from the surface to 5 hPa, corresponding to a vertical resolution of 40-400m in the boundary layer (7 levels) and about 800m around the tropopause. Turbulent diffusion follows the *Louis* [1979] scheme, while the convection scheme (mass-flux type) is that of *Bechtold et al.* [2001]. MOCAGE uses the chemical scheme RACMOBUS, which combines the REPROBUS scheme [*Lefèvre et al.*, 1994] for the stratosphere and the RACM scheme [*Stockwell et al.*, 1997] for the troposphere. RACMOBUS includes 119 individual species, among which 89 are prognostic variables, and 372 chemical reactions. The model also parameterizes dry deposition [*Nho-Kim et al.*, 2004], and wet deposition in convective clouds [*Mari et al.*, 2000] and in stratiform precipitations [*Giorgi and Chamedeis*, 1986]. The model uses emissions taken from *Dentener et al.* [2004]. The model was run for the period June to August 2004 forced with analysis from the Meteo France ARPEGE model [*Courtier*, 1991]. A realistic daily biomass burning emission inventory of North American boreal forest fires [*Pfister et al.*, 2005] was included in the model for this simulation. A detailed evaluation of the model against observations during ICARTT has already been presented in *Bousserez et al.* [2007]. The modeled CO showed good agreement with the measurements during the period of the plume sampling on the 18 July with a peak at 6 km although the gradients are smoothed out due to too much diffusion (see Fig.1).

3. **Diffusive Ensemble Reconstructions**

Turbulent mixing in a strongly stratified atmosphere originates mostly from vertical displacements, translated in the horizontal direction by the natural tracer vertical/horizontal aspect ratio which depends on the ratio between vertical wind shear and horizontal strain.
In order to estimate dispersion or mixing rates in the mid-latitude troposphere we have performed diffusive reconstructions on the same polluted plume on both sides of the Atlantic. The tracer reconstructions were carried out for the Lagrangian match segments of the DC8 and DLR Falcon flights discussed in 2.1.

A range of values of $D_v$ was applied to the vertical displacement of the parcels. CO concentrations were averaged over $N$ parcels arriving along the flight track (with $N=1000$) and thus represent an average over the different origins of the air parcels.

### 3.1. Estimates of $D_v$ in mid-latitude troposphere

On 18 July 2004, the NASA DC8 aircraft flew north into the plume at 7 km and south out of plume at 10 km. The reconstructed CO concentrations with different values of $D_v$ are plotted in the first column of Figure 1 with $D_v$ ranging from $0.1 \, \text{m}^2 \, \text{s}^{-1}$ to $1 \, \text{m}^2 \, \text{s}^{-1}$.

The ensembles are composed of $N = 1000$ particles per point and the time of backward integration is 96 hours. In all cases, the peak of CO mixing ratio associated with the plume is correctly placed around 19:00 UT. The sharpest gradient, and hence the position of the border of the plume, is shifted by about 3 minutes of flight time in the reconstructions with respect to the measurements, which is less than 50 km. This distance is less than half of the horizontal grid size of the advecting wind fields, and errors of this order are likely to result from interpolation errors in horizontal advection. The sharp gradient of mixing ratio is not affected by changes in the diffusivity coefficient $D_v$ of several orders of magnitude (not shown), whereas the mixing ratio at the peak decreases as the value of $D_v$ increases. The best fit is obtained with a value of $D_v$ close to $0.5 \, \text{m}^2 \, \text{s}^{-1}$. Note that the results from all the reconstructions discussed here including this flight much better
agreement with the data than the results from MOCAGE interpolated along the flight tracks. This point is discussed further in section 6.

During the flight on 22 July, the German Aerospace Center (DLR) Falcon encountered a divided Alaskan forest fire plume off the Spanish coast. Lagrangian reconstructions with diffusivities ranging from $10^{-1}$ m$^2$ s$^{-1}$ to 3 m$^2$ s$^{-1}$ are shown in the second column of Figure 1. The peak in CO seen in the profile corresponds to the passage through one remnant of the fire plume. The aircraft entered the plume at 6.5 km and exited it at 2.5 km. The peak and its sharp border are correctly located in the reconstruction, regardless of the the value of $D_v$. The best reconstructed CO profile is obtained with values of $D_v$ of 1 m$^2$ s$^{-1}$.

The third column in Figure 1 shows diffusive reconstructions of the plume remnant on 23 July over France as observed by the DLR Falcon. The sharp gradients observed in the profile on the borders of the plume are associated with sudden changes in direction of the aircraft. These transitions are present in the reconstructions even if the absolute values do not match measurements very well. Two observed CO peaks with 250 ppbv and 200 ppbv, respectively, are reproduced using a $D_v$ of 1 m$^2$ s$^{-1}$. Outside the peaks, values of CO are overestimated, possibly due to initialization inaccuracies or chemical losses, and decrease as $D_v$ increases.

From these reconstructions on both sides of the North Atlantic, $D_v$ is estimated to lie between 0.5 and 1 m$^2$ s$^{-1}$. In the reconstructions on 18 July, large variations in the value of $D_v$ have little impact on the gradient at the edge of the plume, and on 23 July gradients change little with $D_v$. Also, the estimated diffusivity parameter $D_v$ is larger in the case of the Falcon plume sampling compared to the DC8 sampling 5 days earlier. These differences may be related to the fact that the plume is more fragmented after
crossing the Atlantic, and complex spatial patterns favor small-scale mixing. Also, it has been shown that the dynamical situation changed in the middle of the Atlantic during the passage to Europe with a warm conveyor belt moving up from the southwest which was likely to be more turbulent (Real et al. [2007], section 5.3). This resulted in a change in direction in the transport pathway, and is consistent with a larger value of the diffusivity parameter over the eastern Atlantic. Differences in the integration time (4 compared to 8/9 days) were not found to be important in this case (see next subsection).

### 3.2. Uncertainties in the Lagrangian reconstructions

The first important source of error is the representation of advective transport. Small errors in the calculation of the origin position of the parcels may imply large differences in the interpolated mixing ratios from the global model. Even if the model represents accurately the tracer gradients and concentrations in different air masses, inaccurate trajectories could still lead to interpolation of, for example, clean background values instead of plume values and an underestimation in concentrations along the reconstructed flight segment. Adding diffusion prevents spurious fluctuations due to chaos but does not prevent errors due to biases in the analyzed winds. Low dispersion in the cloud of parcels associated with a particular measurement point increases this error, and a whole flight segment may show a fake tracer structure. If dispersion in the ensemble of parcels is large, this effect is reduced. In practice, advection errors depend on the representation of regional and synoptic flow structures in the ECMWF analysis.

As mentioned above, other sources of error could be related to the length of the runs and the number of parcels. It has been shown that after a transient time of the order of 50 hours, the reconstructions are weakly dependent on the length of the run, which
is theoretically justified by the fact that the present method provides a discretization of
the Green’s function of the advective-diffusive equation [Legras et al., 2005; Pisso and
Legras, 2008]. In addition, this forest fire case is an initial value problem and the length
of the run is constrained by the time elapsed between the flights and the emissions. As
for the sensitivity of tracer fluctuations to the size $N$ of the ensemble of parcels, it is
proportional to $N^{-1/2}$. Previous work has shown that, depending on the time interval of
the simulations along the flight tract, results with $N=100, 500, 1000$ give good results in
terms of reproducing of small-scale features [Legras et al., 2005; Pisso and Legras, 2008].
See also Figure 1 in Real et al. [2008].

These results also depend on the interpolated values of CO from the MOCAGE model
4 or 8/9 days before. Model results depend on many factors including injection height
for the forest fire emissions as well as the boundary layer and convective schemes in the
model. In addition, chemical transformations in air masses are not taken into account
in this paper. For CO, we expect this to be reasonably small as shown by Real et al.
[2007]. This is discussed further in the companion paper [Real et al., 2008] together with
the chemical evolution of other species such as $O_3$.

4. Tracer gradient formation, long-range transport and local dynamical
barriers
Lagrangian calculations can provide an accurate description of long-range transport.
The aim of this section is to analyze the precision achieved in the characterization of
the geographical origin and dynamical behavior of the air masses by means of additional
backward trajectory calculations.
Regions of high dispersion in the flow (i.e. rapid separation of parcels) are characterized by maxima in Lyapunov exponents [Pierrehumbert and Yang, 1993] which describe the transformation of an infinitesimal spherical cloud surrounding a particle into an ellipsoid in a local reference frame relative to the parcel.

Non-diffusive ensembles of back trajectories were initialized along the track of 18 July DC8 flight, to investigate the origin of the air masses and to estimate the stirring using the finite 3D Lyapunov exponents (see Benettin et al. [1980]; Legras et al. [2005] for details of method). Figure 2 shows the correlation between sharp tracer gradients, rapid back trajectory separation and peaks in the Lyapunov exponents along the flight tracks.

From the upper panel in Figure 2, the diversity in the origin of air masses measured along the DC8 flight is apparent. A first separation of the backward trajectories occurs within the first two days between a branch remaining at mid-latitudes and another branch rapidly advected northward within a jet streak with winds above 50 m s$^{-1}$ at 300 hPa that extends over northern Canada. This separation is associated with the step in latitude and longitude at time 19.15 UT (in hours from 00:00 the day of the flight) in panels b and c. No step in altitude is associated with this separation as seen from panel d. A step in altitude is produced, however, at a later stage at 18.6 UT and 19.05 UT between parcels remaining near the tropopause, and those advected backward within the updraft south to the entrance of the jet streak. The descending parcels are those which sample the polluted air near its origin as seen in panel e. The reconstructed CO curve fits very well the observations after a shift of 3 minutes along the flight track, that is 50 km in distance, due to the advection errors. This matching indicates that the peak of CO at time 19.1 in
the reconstruction is spurious. As it arises from parcels that stay near the tropopause, it is likely due to an overestimate of vertical transport of pollution by MOCAGE.

The Lyapunov exponents (panel f) exhibit smaller values inside the reconstructed polluted plume than outside, showing that there was low dispersion inside the plume, preserving it for a fairly long time. Extrema of the largest (positive) and smallest (negative) Lyapunov exponents are associated with the edges of the reconstructed plume. Large absolute values are also associated with the first separation in latitude and are seen outside the plume before time 18.6, indicating multiple stretching events in the region. Similar observations and conclusions hold for the case of DLR Falcon flight on 23 July (not shown).

Hence, these results show that fast separation by synoptic-scale transport, marked by peaks in the Lyapunov exponents, is the main cause of steep gradients in the tracer fields. As backward trajectories sampling the plume descend to altitudes of 4 km or lower, there is no need to evoke deep convection penetrating the upper troposphere to explain the observations as suggested by Damoah et al. [2006].

5. Comparison with previous diffusivity estimates

Previous estimates of $D_v$ in the troposphere are limited. [Schumann et al., 1995] made an estimate ($0.6 \text{ m}^2 \text{s}^{-1}$ on average) based on dispersion of aircraft plumes using a Gaussian framework which appear to be consistent with the results presented here. Other estimates have been made for the stratosphere, where $D_v$ varies by several orders of magnitude, from $10^{-2} - 10^{-1} \text{ m}^2 \text{s}^{-1}$ [Balluch and Haynes, 1997; Legras et al., 2005] in the polar region and mid-latitudes to $0.5 \text{ m}^2 \text{s}^{-1}$ in the subtropics [Pisso and Legras, 2008] and so are not directly comparable to results for the troposphere. Estimates of the effect of small-scale unresolved
motion should not be confused with estimates of diffusion obtained by averaging motion over meso-scale and synoptic-scale events [Hegglin et al., 2005]. This is a large-scale limit, in the Taylor sense, which can only match by chance the effect of small-scale diffusion considered here.

In fact, most previous studies focused on the estimation of total or horizontal diffusivity in the troposphere. Since, in stratified flows (in absence of convection), vertical and horizontal diffusion are not independent processes but two aspects of the same phenomenon, it is possible to use typical aspect ratios of tracer structures to establish a correspondence between vertical and horizontal components of the diffusivity tensor. For this reason, we have also reconstructed the CO data for 18 July using horizontal instead of vertical stochastic perturbations. The results with \( D_h = 10^3 \text{ m}^2 \text{ s}^{-1} \), \( D_h = 10^4 \text{ m}^2 \text{ s}^{-1} \), \( D_h = 10^5 \text{ m}^2 \text{ s}^{-1} \), shown in Figure 3 suggest that reconstructions using a value of \( D_h = 10^4 \text{ m}^2 \text{ s}^{-1} \) give the best agreement with the measurements, in this case.

Following Haynes and Anglade [1997] and Young et al. [2007], the square root of the quotient \( D_h/D_v \) yields an estimate of the aspect ratio of tracer structures induced by shear and strain. Using the upper limit \( D_h = 10^4 \text{ m}^2 \text{ s}^{-1} \) and the estimated values \( D_v = 0.5 - 1 \text{ m}^2 \text{ s}^{-1} \) gives aspect ratios in the range of 100 to 140 for this case. These values are of the same order but slightly smaller than the value 200 previously estimated by Haynes and Anglade [1997] for the stratosphere based on the same methods. Our estimates are some of the first for the free troposphere but appear to be consistent with unpublished work based on lidar data analysis (F. Ravetta, pers. comm.).

The results presented here for the free troposphere can be compared with previous estimates of \( D_h = 1 - 5 \times 10^4 \text{ m}^2 \text{ s}^{-1} \) [Gifford, 1982; Sillman et al., 1990; Mauzerall et al.,...
These values are somewhat larger than the estimates from our study and were determined for plumes pollution plumes close to the source regions in or near the boundary layer. In these cases, mixing was likely to have been governed by small-scale turbulence with little influence from large-scale advection. This is in contrast to the fire plume case studied here where transport was governed by synoptic advection and dispersion by the strain, which is measured by the Lyapunov exponent of the flow, rather than diffusion. For comparison, smaller values of $5 \times 10^3 \text{ m}^2 \text{ s}^{-1}$ were obtained by [Waugh et al., 1997] for the stratosphere which can be expected given the higher stability in this region of the atmosphere.

Turbulent diffusion smooths out small-scale structures created by the dynamics, limiting the strength of the gradients and the size of the filaments. In the simple case of a blob close to a hyperbolic point, the length of the blob evolves according to $e^{\lambda t}$ but the width cannot stretch under a size of the order of $\sqrt{D/\lambda}$ because of the equilibrium between strain and diffusion. Hence, the tracer concentration diminishes as $e^{-\lambda t}$ inversely to the length growth rate. Here, diffusion is essential in fixing the width of the blob but the damping rate of the concentration does not depend on it, unlike a common assumption made in chemical models. In real flows, the relation between stretching and decay rate is somewhat more complicated because shear dominated flows, like in the atmosphere, are much less efficient at stirring and mixing than idealized pure strain and the Lyapunov exponent is only an upper bound of the decay rate but nevertheless the mixing properties are fairly independent of the underlying small-scale diffusion [Shuckburgh and Haynes, 2003; d’Ovidio et al., 2008].
Mixing is often represented with exponential decay rates of tracer mixing ratio to background concentrations ($\tau^{-1}$), which for the case examined here have been found to range between 0.1 and 0.2 day$^{-1}$ [Real et al., 2007; Arnold et al., 2007]. As small scale mixing is controlled by turbulent motions, these exponential decay rates can be compared directly to the Lyapunov exponents which exhibit values ranging from 0.1 day$^{-1}$ inside the smoke plume to 1 day$^{-1}$ outside as seen in Fig. 2, panel f. It corresponds to the DC8 transect, values for the Falcon transect are of the same order of magnitude and are also consistent with previous estimates [Real et al., 2007; Arnold et al., 2007]. The latter are pointwise estimations and large variability can be expected from the values of strain inside a folded air mass like the one studied here. In another approach, developed for the Chemical Lagrangian Model of the Stratosphere (CLaMS) [McKenna et al., 2002], mixing is controlled by the local horizontal strain and vertical shear rates and further decomposed in directions parallel and transverse to the flow. The numerical parameters $D_\pm = r_0 e^{\pm 2\lambda \Delta t}$ (McKenna et al. [2002], appendix B) representing the horizontal components of the diffusivity could be estimated from the values of Lyapunov exponents for our case (between 0.1 day$^{-1}$ and 1 day$^{-1}$) depending on the spatial and temporal scales $r_0$ (initial separation of particles) and $\Delta t$ (time step) but would require a specific CLaMS model run.

6. Relation to Eulerian global models

In this section, we relate the estimates of diffusivity obtained in this study to the “diffusion” in global chemical models in an attempt to estimate the spatial resolutions required to accurately simulate pollutant plume transport and dispersion in the free troposphere.

Increasingly sophisticated advection schemes have been developed since the seminal work of Van Leer [1977] intending to preserve tracer gradients such as the slope preserving
scheme [Russell and Lerner, 1981], second order moments [Prather, 1986] or different forms of semi-Lagrangian advection with shape preserving interpolation [Williamson and Rasch, 1989], used in MOCAGE [Josse et al., 2004]. Although numerical diffusion is reduced, if these methods are used to integrate a pure transport equation, the result (if a suitable mass conserving equation is satisfied) will be, in practice, the solution of an advection-diffusion problem, with a numerical coefficient depending on the method, the time and space resolution used in a particular run etc. Contrary to CLaMS, in finite volume methods the numerical diffusion depends on the mean wind which is less physically justified.

In general, global representation of horizontal diffusion is neglected in Eulerian models since they are already too diffusive at current horizontal resolutions. Available computing resources force global models to be run at resolutions that prevent fine scale structures being fully resolved. In this case, the “effective” diffusivity resulting from the numerical scheme in a global model is larger than the “physical” turbulent diffusivity corresponding to a certain grid scale of the wind [Legras et al., 2003; Pisso and Legras, 2008]. Using reverse domain filling techniques for tracer reconstruction and comparison with the SLIM-CAT model in the lower stratosphere, Tan et al. [1998] estimated an horizontal diffusivity of the order of $10^6 - 10^7 \text{ m}^2 \text{s}^{-1}$ in coarse resolution Eulerian models. We have found a value for $D_h$ of the order of $10^4 \text{ m}^2 \text{s}^{-1}$ based on measurements (see also [Tan et al., 1998] table 1).

Although a general formula for the dependence of effective numerical diffusivity has to take into account the setting of a particular model, we can obtain an order of magnitude estimate for the global resolution ($2^\circ \times 2^\circ$) of MOCAGE by comparing the interpolated global
model results with diffusive reconstructions for the DC8 flight. The value of $1 \text{m}^2 \text{s}^{-1}$ for vertical perturbations is still too low to match the MOCAGE results interpolated along the flight tracks (Fig. 1). It can be seen from Fig. 3 that the global model results are equivalent to applying a horizontal diffusivity of $10^5 \text{m}^2 \text{s}^{-1}$ or more which is greater than the $10^3 - 10^4 \text{m}^2 \text{s}^{-1}$ based on the our stochastic reconstructions and which give the best agreement with the data. In other words, the "natural" value of real horizontal turbulent diffusion is smaller than the value in models related to their numerical schemes at current model resolutions. Notice that large-scale models usually use the same resolution for dynamical and tracer fields. Our results suggest that a significant improvement could be to increase the resolution of the tracer fields alone.

On the other hand, numerical diffusion is explicitly represented in meso-scale models where diffusion is defined as a function of the scale of spurious numerical waves. Several parametrizations based on dynamical parameters exist. For example, the Smagorinsky deformation closure can be used to estimate the resolution needed to represent the effective $D_H$.

As an example, the horizontal diffusivity in MM5 [Grell et al., 1995] is written as

$$D_h = D_{h0} + \frac{1}{2} k^2 \Delta x^2 S$$

where $k = 0.4$ is the von Karman constant, $S = [(\frac{\partial u}{\partial x} - \frac{\partial v}{\partial y})^2 + (\frac{\partial v}{\partial x} + \frac{\partial u}{\partial y})^2]^{1/2}$ represents shear induced diffusion [Smagorinski, 1963] and $D_{h0} = K \frac{\Delta x^2}{\Delta t}$ is the basic diffusivity related to the model resolution in space and time, needed in meso-scale models to control nonlinear instability and numerical aliasing. The constant $K$ is user defined [Xu et al., 2000] and its default value in MM5 is $3 \times 10^{-3}$ [Grell et al., 1995].
The order of magnitude of the Smagorinski closure can be approximated by using the Lyapunov exponents calculated for this study by approximating the shear with the Lyapunov exponent $S \approx \lambda^{-1}$. The relation between the spatial and time resolution that is achieved for a certain diffusivity is given by:

$$\Delta x = \sqrt{D_h[3 \times 10^{-3} + 2k^2\lambda^{-1}\Delta t]}$$

These calculations suggest (Fig. 4) that global CTMs with a dynamical time step of 15 or 30 minutes require horizontal grids smaller than 40 km to reach the values of $D_H$ estimated in this study.

The optimal vertical/horizontal balance in spatial resolution is also important. The values of $D_v$ and $D_h$ obtained in section 5 were used to estimate the vertical/horizontal aspect ratio between 100-140 for the case of forest fire plume dispersion in the free troposphere. Studies of ozone vertical profiles have shown that such layers occupy a significant part of the troposphere [Newell et al., 1999] and the median thickness is around 500 m [Thouret et al., 2000, 2001]. Therefore, it is important to quantify their characteristics and processes governing their formation, maintenance (of gradients) and eventual dispersion.

The actual vertical and horizontal resolutions applied in the MOCAGE runs used here (800 m vertical and 220 km horizontal) lead to an aspect ratio of 1:275. The results presented here suggest that in order to capture long-range transport of pollutant plumes, global models need to be run at vertical resolutions of the order of 500 m or less and horizontal resolutions of less than 40 km. At these resolutions, it may also be necessary to include parametrizations for horizontal and vertical diffusion in such models. Whilst previous studies of the impact of resolution on photochemistry showed rather small effects down to 110 km (e.g. for example Wild and Prather), results presented in the companion
paper, Real et al. [2008], show this is not always the case and that significant errors can occur.

7. Conclusions and outlook

We have studied the dynamical evolution of a forest fire plume during long-range transport in order to quantify mixing and dispersion in the free troposphere. Stochastic Lagrangian reconstructions were performed at different stages of the plume originated in Alaska and transported across the Atlantic. The present work focused on different dynamical processes acting during long range transport controlling the tracer gradients enhancement and decay. In a companion study, the chemical transformations within the plume are also taken into account.

This is the first time the backward Lagrangian diffusive ensembles have been applied in the free troposphere. The use of analysed meteorological winds yielded a small shift in the location of gradients in the reconstructed profiles. Also, interestingly, explicit convective parameterizations in the trajectory calculation were not required in the calculation of vertical displacements with significant vertical motion taking place due to the information in the analyses alone.

We have estimated vertical and horizontal turbulent diffusivity coefficients, $D_v$ and $D_h$, based on stochastic reconstructions of CO along matching flight segments across the plume. The vertical turbulent diffusivity $D_v$ ranges between 0.5 and $1 \text{ m}^2 \text{ s}^{-1}$ and the horizontal diffusion $D_h$ upper bound is of the order of $10^4 \text{ m}^2 \text{ s}^{-1}$. These values represent sub-grid processes in the standard spatial resolution of operational centers like ECMWF and are consistent with previous, albeit rather limited, estimates of $D_h$ and $D_v$. Exponential decay (mixing to background) is often used to represent mixing in photochemical
trajectory models. Although turbulent diffusion contributes when filaments produced by shear reach the diffusive scale, in general the decay rate is controlled by the strain itself. The Lyapunov exponents, a measure of the strain, estimated inside the plume range between 0.1 and 1 day$^{-1}$, which is consistent with exponential decay rates between 6 and 10 days found in previous studies. The estimates of the horizontal and the vertical components of the diffusivity tensor yield estimates of the aspect ratio of tropospheric tracer structures of the order of 100—150.

Global chemical models contain inherent numerical diffusion related to coarse resolution. Here, we estimate that in the case of the MOCAGE model, this diffusion is larger than $10^5 m^2/s$, at least one order of magnitude larger than the value estimated from the reconstructions of the in-situ measurements. We have estimated that the resolution required to be able to simulate long-range transport of pollutant plumes in the free troposphere and their dispersion is less than 40 km in the horizontal and 500 m in the vertical. These values provide an upper bound which will need to be refined once global models are run at sufficient resolutions with suitable parameterizations for horizontal and vertical diffusion. Note that, here we only consider resolutions required for accurate dynamical transport of pollutant plumes in the free troposphere where advection is dominant. Non-linearities in the chemistry, particularly at plume edges, may require even higher resolutions (see [Real et al., 2008]).

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Figure 1. The blue curves represent CO mixing ratios reconstructed using ensembles of diffusive backward trajectories as a function of flight time. First column DC8 flight on 18 July. Second and third columns, Falcon flights on 22 and 23 July, respectively. The in-situ measurements are plotted in red and MOCAGE CTM interpolated CO in black in every panel. In all figures vertical axis represents CO mixing ratio in ppbv and horizontal axis represents flight time (hrs). Rows correspond to values of $D_v$ ranging from $0.1 \text{ m}^2 \text{ s}^{-1}$ to $10 \text{ m}^2 \text{ s}^{-1}$ depending on the flight.
Figure 2. In panel (a) non-diffusive back trajectories released along 18 July DC8 flight track are plotted in grey. Colors indicate time of the previous positions of the parcels counted backward from 19 July, 2004 at 12:00UT. Longitude, latitude and altitude of the backward positions of the parcels along trajectories are plotted in panels (b,c,d) using the same color code as in panel (a), and as a function of release time along the flight track. CO measurements (blue) and CO reconstructions for $D = 0.5 \text{ m}^2 \text{ s}^{-1}$ (black) are shown in panel (e). Three-dimensional Lyapunov exponents calculated over a five day interval are plotted in panel (f).
Figure 3. Diffusive reconstructions of DC8 CO with horizontal instead of vertical stochastic perturbations. Black points correspond to in-situ CO data, the black dashed line represents the direct interpolation of MOCAGE CO along the flight track at 0:00UT. Blue, green and red lines represent diffusive ensemble reconstructions with purely horizontal stochastic perturbations corresponding to diffusivities $D_h$ of $10^3 \text{ m}^2 \text{ s}^{-1}$, $10^4 \text{ m}^2 \text{ s}^{-1}$ and $10^5 \text{ m}^2 \text{ s}^{-1}$, respectively.
Figure 4. Relationship between time and space steps corresponding to different values of diffusivity estimated using a meso-scale (MM5) model setup. MOCAGE parameters place its diffusivity in the range of $3 \times 10^5 \text{m}^2 \text{s}^{-1}$. See text for details.