# Long Range Transport and Air Quality

## **Group H**

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## 1. Introduction

Air quality and pollution are typically treated as a local issues. However, since the advent of satellite observations, there has been increasing evidence of their regional and global nature. It is now recognized that long range transport of pollutants across national boundaries and continents can carry pollutants far away from their sources. Thus, local air quality can be impacted by pollution generated elsewhere, to the extent that critical levels may be exceeded. For example, the average concentration of ozone in remote parts of East Asia has already reached levels where it can jeopardize agricultural and natural ecosystems there. Also, air quality and climate are closely coupled through various atmospheric processes. Aerosols have a direct role in radiative forcing, while certain chemical species like CO and  $NO_x$  influence the formation and lifetimes of several greenhouse gases. In turn, meteorological parameters can affect the sources, transport, transformations and deposition of pollutants. A review of various aspects of global air quality and pollution is given, for example, by Akimoto (2003).

There are several aspects of the problem of long range transport of pollutants that could be of interest. The timescales involved in transport are crucial. Timescales for inter-continental transport are of the order of 3 to 30 days. Hence, it is typically of relevance for species having a lifetime within this range, though shorter lived species may be influenced by reactions with longer lived compounds. Another aspect involves understanding the typical pathways of transport. One could also investigate the temporal patterns of long range transport, including seasonal and inter-annual variability, associated for example, with El Niño. Finally, it is important to predict the change in the patterns of transport due to climate change, or due to actual or simulated changes in anthropogenic activities.

In this report, outputs from the model FLEXTRA, as well as results from the model FLEXPART that have been reported in the literature, are used to study some of the aspects of long range transport of pollutants in the atmosphere. These models will now be described in some detail, before moving on to the study per se.

### **1.1 Transport models**

Apart from analysis of observational data, there are many theoretical tools to study pollutant transport. Some of the simplest are the trajectory models. In such models, a small volume of air, called a particle, is advected using the mean horizontal and vertical winds from a meteorological model. Examples of such models include HYSPLIT (Draxler and Rolph, 2003 and Draxler, 2003), FLEXTRA (Stohl *et. al.* 1995, Stohl and Seibert, 1998), LAGRANTO (Wernli and Davies, 1997) and TRAJKS (Scheele *et. al.* 1996). These models are often run backwards in time beginning from a given location, resulting in the so-called 'back-trajectories'. These indicate where the tracer particle came from, and are useful to discern pollution sources and for interpreting in-situ measurements.

However, simple trajectory models suffer from some serious drawbacks. Trajectories are paths of small parcels or particles that maintain their integrity and do not mix with the ambient. In contrast, measurements involve a finite and typically large volume of air. Remote measurements such as LIDAR require finite backscatter volumes, while in-situ measurements also involve large volumes because of sampling times. Non-uniformities in the flow, typical of the atmosphere, will deform a spherical volume into an elongated structure. By a process known as stirring (Haynes 2003), over the course of time, repeated stretching and wrapping of fluid

elements splits an initially compact volume into filamentary structures which may be spread over considerably large areas. Such processes can be shown to apply to back-trajectories as well, and is the reason why a single trajectory cannot be representative of even a small sampling volume. Single trajectories also cannot take into account the turbulence in the atmospheric boundary layer, which causes the rapid dispersion of air masses, especially in the initial stages, which in turn amplifies the stirring processes.

These issues can be better handled by eulerian Chemistry Transport Models (CTMs) and Lagrangian Particle Dispersion Models (LPDMs). These can also be run in the receptor oriented mode (backward in time). The CTMs essentially solve the advection-diffusion equation on an Eulerian grid. They can however suffer from finite grid resolution (for example, causing instantaneous mixing within a grid), and numerical diffusion, which may be comparable to the actual diffusion. LPDMs, for example FLEXPART, account for stirring and turbulence by calculating the trajectories of a large number of particles transported by the mean wind, as well as by stochastically modeled turbulent fluctuations. However, output from a LPDM run consists of four dimensional gridded data and is thus much more complex than a simple trajectory. Hence special visualization and analysis techniques are necessary for interpreting the output from a LPDM.

### **1.2 FLEXTRA and FLEXPART**

FLEXTRA is a simple trajectory model that calculates air mass trajectories using meteorological data provided by ECMWF (European Centre for Medium Range Weather Forecasts). Backward trajectory data and an interactive plotter is available on the web at <u>http://tarantula.nilu.no/trajectories</u>. The data and plots are available for four times in a day, from 1996 until the present date minus three days, for over 290 locations. A typical plot for the observation station of Bukit Kototabang (0.20°S 100.32°E), situated on the island of Sumatra, Indonesia is shown in Fig.1. These represent trajectories arriving at this station at 18:00 hrs UTC



Fig. 1: A typical FLEXTRA back trajectory plot, shown here for receptor location of Bukit Kototabang, Indonesia (0.20°S 100.32°E), for arrival time 18 UTC on 6 June 2000. See text for more details.



Fig. 2: The column integrated potential emission sensitivity (PES) plot for a backward FLEXPART simulation at Zeppelin, Norway (78.90°N 11.88°E). The PES value in a given grid cell is a measure for the mixing ratio at the receptor (in this case, Zeppelin), that a source of unit strength (1 kg/s) in that grid cell would produce. Here, the PES is integrated over the entire vertical column.

on 6 June 2000. To give some idea of the dispersion of air volumes, three trajectories are available, each representing a different height of arrival (indicated by the legends in the upper right corner). Each 3 hour interval along a given trajectory path is indicated by a small legend and each 24 hour interval by a big legend. Variation of height along a given trajectory is indicated by color coding, whose scale is depicted along the bottom of the plot. The accuracy of trajectories has been estimated to be typically around 20% of the travel distance, though larger errors (of the order of 100%) are possible for individual cases (Stohl and Seibert, 1998).

As mentioned, FLEXPART (Stohl *et. al.*, 2002, 2005) is a LPDM that releases a large number of particles over the sampling time at the desired location, and calculates their trajectories using stochastically modeled turbulent fluctuations superposed on the grid scale winds provided by ECMWF. In the backward mode, the cloud of particles is called a retroplume. It is also equipped with a scheme for moist convective transport. FLEXPART is freeware and is currently being used by many groups all over the world (<u>http://transport.nilu.no/flexpart</u>). However, there is also an internet based front end for displaying the outputs from FLEXPART, is available for a large number of observation campaigns and research sites (<u>http://transport.nilu.no/products/interactive-tool</u>). There are several ways of visualizing the output and some of these output products (for backward evolution) are available via the internet





Fig. 3: View from the Zeppelin station under clear conditions on 26 April, and during the smoke episode on 2 May 2006.

based front end. One of them is the 'potential emission sensitivity' distribution (PES). The value of the PES (in units of s kg<sup>-1</sup>) in a particular grid cell is proportional to the particle residence time in that cell. For a backward simulation, it is a measure for the simulated mixing ratio at the receptor, that a source of unit strength (1 kg s<sup>-1</sup>) in the respective grid cell would produce. This can be integrated over the entire vertical column (column-integrated PES) or over the lowest 100 m (footprint PES). The footprint PES is clearly more relevant for anthropogenic emissions. Fig. 2 shows an example of a column-integrated PES for a sampling period of 3 hours ending at 15 hrs UTC, on 1 May, 2006, for the site of Zeppelin in Norway (78.90°N 11.88°E). The receptor point (Zeppelin) is indicated by an asterisk. The numbers plotted on top of the contours are the daily retro-plume centroid positions, and are the closest equivalent of the back-trajectories in the simple trajectory models. To understand contribution from anthropogenic sources, the PES footprint may be multiplied with emission flux densities (in units of kg m<sup>-2</sup> s<sup>-1</sup>) taken from an appropriate emission inventory, resulting in what is called the 'potential source contribution' (in units of ppbv/m<sup>2</sup>).

## 2. Examples of long range transport of pollutants

As mentioned, there is now considerable evidence for the long range transport of pollutants across national boundaries and continents. This is illustrated in this section by a couple of well documented examples.

### 2.1 An Arctic pollution episode

An arctic pollution episode, recorded at the research station Zeppelin (11.9° E, 78.9° N, 478 m a.s.l.), on the western coast of Spitsbergen, Norway, is now considered. Record high levels of air pollution were measured there in the months of April and May, 2006, with the most severe episodes occurring on 27 April and 2, 3 May. Fig. 3 shows views from the Zeppelin station under clear conditions and during the smoke episode on 2 May, 2006. Fig. 4a shows the



Fig. 4: (a) Time series of the daily mean number concentrations of accumulation mode (100–500 nm diameter) particles at Zeppelin for the period April–May 2006, showing a peak on 27 April as well as on 2 and 3 May. Also shown are the median and the 95-percentile for the months of April and May in the years 2000–2005. (b) Time series of equivalent black carbon (EBC) and CO measured at Zeppelin from 24 April to 10 May 2006. Also shown are elemental carbon (EC) and organic carbon (OC) concentrations from weekly samples and daily mean particle mass (PM) concentrations derived from the DMPS (Differential Particle Mobility Sizer) data.



Fig. 5: (a) FLEXPART footprint PES from a backward simulation, and (b) FLEXTRA back-trajectories, both shown for Zeppelin, for the arrival time 0 UTC, 3 May 2006. The approximate region of where the forest fires occurred (taken from satellite data) is indicated by a gray oval. The FLEXPART retroplume centroid positions closely correspond to the FLEXTRA trajectories. It can be seen that the air arriving at Zeppelin passed over the region of forest fires 2-4 days prior to arrival, bringing with it biomass burning emissions.

timeseries of the daily mean number concentration of accumulation mode particles measured at Zeppelin around this period. Fig. 4b shows concentrations of carbonaceous particles and CO measured at Zeppelin, again showing peaks on 27 April and 2 and 3 May.

To understand this pollution episode, we look at backward simulation outputs from FLEXTRA and FLEXPART. Fig. 5a shows the footprint PES from a backward FLEXPART simulation and Fig. 5b shows the FLEXTRA back-trajectories, both for arrival at 0 UTC, 3 May 2006, at Zeppelin. Note that FLEXPART retroplume centroid positions closely correspond to the FLEXTRA back-trajectories. During this period there were a large number of agricultural fires in the Baltic countries, western Russia, Belarus and Ukraine. These were detected by satellites using the MODIS instruments, with more than 300 fires per day detected between 25 April and 6 May 2006. The approximate region of the fires is shown in Fig. 5a and 5b by the gray ovals. Clearly, the air masses arriving at Zeppelin travelled over these regions 2-4 days before arrival, bringing the biomass burning emissions with them. The FLEXPART outputs were in good agreement with satellite observations. For example, the column tracer from a FLEXPART forward simulation agreed very well with MODIS aerosol optical depth fields, and AIRS CO retrievals (Stohl *et. al.* 2007). In this episode, pollutants were transported over several hundred kilometers over a few days.



Fig. 6: (a) Total CO tracer columns for 4, 6 and 9 August 1998, from a FLEXPART forward simulation showing the transport of forest fire emissions from Canada to Europe (b) FLEXTRA back trajectories at Leipzig for arrival at 12 UTC on 9 August, showing a good correspondence with the FLEXPART result (bottommost panel).

#### **2.2 Trans-Atlantic transport of boreal forest fire emissions**

As an example of transport over longer distances, we consider the trans-Atlantic transport of boreal forest fire emissions from Canada to Europe in August 1998. In August 1998, severe forest fires occurred in many parts of Canada, with more than  $10^6$  hectares of forest burnt in the week from August 5 to 11. CO tracer columns from a FLEXPART forward simulation, shown in Fig. 6, show the transport of the forest fire emissions to Europe (Forster *et. al.* 2001). This can also be seen in the FLEXTRA back-trajectories at Leipzig, also shown in Fig. 6. The air masses travelled a distance of well over 5000 km in about a week. Indeed, haze layers and enhanced CO concentrations were found over many parts of Europe in August 1998. Haze layers were observed over Germany, with a peak around 9 and 10 August. The vertical aerosol concentration profiles obtained from FLEXPART were found to be in good qualitative agreement with lidar measurements at various stations in Germany (including Leipzig). It is not possible to obtain vertical profile information using FLEXTRA.

## **3.** Time variability of transport patterns

This section looks at time variability of transport patterns. In particular, intra-annual and inter-annual changes in transport patterns at a given location are considered. The location chosen here is Bukit Kototabang (0.20°S 100.32°E), on the island of Sumatra, in Indonesia.

## **3.1 Intra-annual variability**

As an example of intra-annual variation, the back-trajectories (at Bukit Kototabang) for the months of January and July are compared. This is done by taking various FLEXTRA backtrajectories for January (i.e trajectories ending at randomly chosen time, date and year, but the month being January), and comparing them with various trajectories for the month of July. Fig. 7 shows typical trajectories for these two months. It can be seen that the trajectories for January come in from the North-East while those in July are directed from the South-East. A comparison of large number of trajectories for these two months shows a considerable variation, but the trend of trajectories flowing in from the North in January, and from the South in July is evident. This is consistent with the position of the ITCZ (Inter-Tropical Convergence Zone), which is to the south of Bukit Kototabang in January, and to its north in July. The average position of the ICTZ for these months is indicated by thick black curves in Fig. 7. A more powerful way to investigate such variability is by considering composite plots from FLEXPART outputs, which may be obtained by averaging the PES over the required period.

#### 3.2 Inter-annual variability associated with El-Niño

Next, an attempt was made to look at any inter-annual variability associated with the El Niño or ENSO. First of all, the El Niño years were identified. Now, different criteria are used to identify an El Niño year, like the Southern Oscillation Index (SOI), SST anomalies, etc. Widely used lists are provided by the Western Region Climate Center, the Climate Diagnostics Center,



Fig. 7: Back-trajectories for Bukit Kototabang  $(0.20^{\circ}\text{S}\ 100.32^{\circ}\text{E})$ , for an arbitrarily picked date in January(a) and July(b). Also shown is the average position of the ITCZ for these months. An examination of a large number of trajectories for these months shows that, though there is considerable variation in the length and direction of the trajectories, there is a clear pattern of flow towards the ITCZ, being southwards in January, and northwards in July.

the Climate Prediction Center and the multivariate ENSO index provided by the Climate Diagnostics Center. For the present purpose, a year was taken to be an El Niño or La Niña year only if it appeared on at least three of the above mentioned lists. Then, as for the case intraannual variability, FLEXTRA back-trajectories for El Niño years were compared with those from La Niña and normal years. Such comparisons (El Niño versus La Niña and normal years) were also carried out for the restricted period of the months from November to February, as the effect of El Niño is usually strongest for those months. There appeared to be some trend. For example, trajectories for the month of January in the normal years typically came in from the North (as discussed in the section on intra-annual variability above), while there appeared more uncertainty and variability in direction of the trajectories for the El Niño years. But such a trend could not be unequivocally established, as there were also considerable variations within the El Niño, La Niña or normal years. There are several possible reasons for this. One is that single trajectories are not an useful way to study such trends, and as before, composite plots (for the El Niño, La Niña and normal years) obtained using FLEXPART would be a better way to investigate such patterns. Also, the period (1996-2009) for which FLEXTRA plots are available, is a limited period, with only 1 strong El Niño event and a couple of weaker ones. Finally, the region around Indonesia exhibits a great deal of complexity, with, for example, the Indian Ocean Dipole also having an influence.

## 4. Extreme transport phenomena

#### 4.1 Rapid transport associated with a meteorological bomb

Intercontinental transport typically occurs over timescales of about a few days to a month. Hence it is not considered relevant for species with lifetimes of few hours or days. An



Fig. 8: (a) Total vertical columns of FLEXPART NOx tracer for 9 and 10 November 2001, showing rapid transport across the North Atlantic (less than a day) (b) FLEXTRA back-trajectories at Leipzig for 0 UTC 11 Nov 2001, showing the same.

important example is that of the nitrogen oxides  $(NO_x)$ , which are critical for the formation of ozone in the troposphere, but have lifetimes of the order of hours in the atmospheric boundary layer, and a few days in the troposphere. However, there can be events involving very rapid transport (the so called inter-continental pollution express highways) which can result in inter-continental transport of such short lived species. Such rapid transport can occur with jet streaks in the upper troposphere or due to explosive development of extra-tropical cyclones (meteorological bombs). During their life cycle, such meteorological bombs can develop extremely low core sea-level pressures with the attendant extreme horizontal pressure gradients and surface winds, which can result in rapid transport.

Here, we look at an example of transport associated with a meteorological bomb that caused rapid transport across the North Atlantic, causing anthropogenic NO<sub>x</sub> from North American sources to be transported to Europe (Stohl *et. al.* 2003). A series of developments led to the generation of a bomb just off the east coast of Canada (roughly 60°W, 50°N) at around 0 UTC on November 8, 2001. There was a strong zonal flow south of the bomb centre which formed the express highway for transport. The FLEXPART output shown in Fig. 8a indicates total vertical columns of NO<sub>x</sub> tracer (averaged for 1 hour) ending at 15 UTC on 9 November and 11 UTC on 10 November. The FLEXPART results show that the leading tip of the NO<sub>x</sub> tracer filament had travelled from south of Greenland to Scandinavia, a distance of around 3000 km, in about 20 hours! More than 1 ppbv of NO<sub>x</sub> , a significant amount to influence ozone production, was transported to Europe from North America. The results from FLEXPART are in good agreement with NO<sub>2</sub> retrievals by GOME (the Global Ozone Monitoring Experiment). Shown in Fig. 8b is a FLEXTRA back trajectory ending at Leipzig on 11 August. Again, the

rapid transport across the North Atlantic on the previous day (10 August) can be seen. It must be noted that meteorological bombs are not that uncommon, with Lim and Simmonds (2002) finding 46 and 26 bombs per year in the Northern and Southern Hemisphere, which is probably an underestimate. These authors also report a significant upward trend of bomb occurrence during the last two decades of the twentieth century, which may be related to global warming.

#### 4.2 Weak transport in the Santiago basin

From the case of very rapid transport across large distances, we now move to the other extreme. In Fig. 9, typical back-trajectories for Santiago, Chile (33.50°S 70.67°W) are shown, for randomly picked arrival times. An examination of a large number of such trajectories (with randomly selected time, date and year of arrival) indicates that a large proportion are like those shown in Fig. 9, being highly localized and confined to a small area. Indeed, the average wind speeds in this region (less than 2 m/s) are much lower than the global average. All this suggests the possibility that the outflows from this region could also be low. This turns out to be true. Santiago is situated in a valley, with low winds speeds and frequent thermal inversions, which restrict the dispersion of pollutants. It is thus not surprising that, even with emission levels comparable to several other cities, Santiago currently ranks as one of the most polluted cities in the world, and frequently confronts pollution emergencies (Romero *et. al.* 1999, Schmitz 2005).

The FLEXTRA trajectories are calculated with wind fields provided by ECMWF, which have rather coarse latitude/longitude resolution of  $1.125 \times 1.125$  (~ 100 km at Santiago). Thus, whilst the trajectories seem to be consistent with trapping in the valley, this is quite surprising as it is not clear that the trajectories will include information about the wind field at small scales.



Fig. 9: Arbitrarily selected back-trajectories for Santiago, Chile (33.50°S 70.67°W). A majority of the large number of randomly selected trajectories were like those shown here, being highly localized and extending over relatively short distances.

## **5.** Conclusions

This study was basically concerned with using outputs from a couple of transport models to study the long range transport of pollutants in the atmosphere. After an introduction to the scope and relevance of atmospheric transport, some of the theoretical tools used to study long range pollutant transport are described. One of these is the simple trajectory model, where a tracer particle is advected using meteorological data. These are often evolved backward in time from a given location in order to discern sources of pollution. An example of such a model is FLEXTRA, which is described in detail, as outputs from this model are used in this study. However, simple trajectory models cannot account for the stirring processes in the atmosphere, which result in the filamentation of air elements, and turbulence in the atmospheric boundary layer. These are better taken into account by Lagrangian Particle Dispersion Models (LPDMs), which release a large number of particles over a period of time, which are transported by the mean wind as well as stochastically modeled turbulent fluctuations. These are also often run in the backward time evolution mode. Outputs from a LPDM called FLEXPART are also used here. Hence, the model, as well as interpretation of its output is discussed at some length.

Some well documented examples were used to illustrate the global nature of air quality and pollutant transport. For example, emissions from forest fires were transported across the Atlantic, from Canada to Europe in August 1998. The vertical aerosol concentration profiles obtained from FLEXPART forward simulations were found to be in good qualitative agreement with lidar measurements at various stations in Germany.

Temporal variations in transport patterns were investigated by considering some aspects of the intra-annual and inter-annual variability of simple backward trajectories at a given location. However, composite FLEXPART plots are more suited for such a study.

Finally, some examples of extreme transport are discussed. One of these involved rapid transport associated with explosive cyclogenesis, which resulted in transport across the North Atlantic in about a day.

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