ATMOSPHERIC DISPERSION MODELLING OF PARTICULATE AND GASEOUS POLLUTANTS AFFECTING THE TRANS-MANCHE REGION

Stylianos Plainiotis

A thesis submitted for the degree of
Doctor of Philosophy at
The University of Greenwich

December 2006

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Declaration

I certify that this work has not been accepted in substance for any degree, and is not concurrently submitted for any degree other than of Doctor of Philosophy (Ph.D) of the University of Greenwich. I also declare that this work is the result of my own investigations except where otherwise stated.

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List of selected publications produced by the author during candidature

Conference Papers:

1. S Plainiotis, K A Pericleous, B E A Fisher: Quantifying the input of wind blown dust to southern European cities. Proceedings of the 6th International Conference on Urban Air Quality Limassol, Cyprus, 27-29 March 2007

2. K.A. Pericleous, S. Plainiotis, and B.E.A. Fisher, 2006: Airborne Transport of Saharan Dust to the Mediterranean and to the Atlantic. Proceedings of the Second IASTED International Conference in Environmental Modelling and Simulation, Acta Press ISBN 0-88986-617-1

3. S. Plainiotis, K.A. Pericleous, B.E.A. Fisher, and L. Shier, 2005: Application of Lagrangian Particle Dispersion models to air quality assessment in the Trans-Manche region of Nord-Pas-de-Calais (France) and Kent (Great Britain). Proceedings of the 10^{th} Int. Conf. on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes, Sissi, Crete, October 17-20, 2005, pp. 398-403

4. S. Plainiotis, K.A. Pericleous, B.E.A. Fisher, and L. Shier, 2005: Forward and Inverse Transport of Particulate Matter and Gaseous Pollutants Affecting the Region Bordering the English Channel, Proceedings of the 16^{th} IASTED International Conference on Modelling and Simulation (MS-2005), pp.164-169, Acta Press pp.459-090

5. S. Plainiotis, K.A. Pericleous, B.E.A. Fisher, and L. Shier, 2005: Modelling high particulate matter and ozone episodes in the Trans-Manche region. Proceedings of the 5th International conference on Urban Air Quality 5, 2005

Books:

Y. Ji and S. Plainiotis (09-2006): Design for Sustainability. Beijing: China Architecture and Building Press. ISBN 7-112-08390-7

Abstract

This thesis describes the development of a methodology to determine large-scale and meso-scale atmospheric dispersion patterns. The research is only concerned with outdoor exposure to atmospheric pollutants and aims to identify pollution sources using dispersion modelling with the assistance of ground level measurements from British, French and other monitoring stations and remote sensing technology.

Lagrangian Particle Dispersion (LPD) models compute trajectories of a large number of notional particles and can be used to numerically simulate the dispersion of a pollutant (passive tracer) in the planetary boundary layer. Two widely used atmospheric dispersion models were employed: the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model by R. Draxler, and the model FLEXPART by Stohl et al. Both models possess forward tracking and inverse (or receptor-based) modes. Meteorological data output from the PSU/NCAR Mesoscale model (known as MM5), or datasets from the European Centre of Medium-range Weather Forecast (ECMWF) are used to drive the dispersion models. Linkage routines were developed to interpret the LPD codes with the required meteorological information.

This study aims to determine whether current approaches and practice for atmospheric dispersion modelling are reliable, consistent and up-to-date. An inter-comparison of the models FLEXPART and HYSPLIT is performed for known episodes to determine their accuracy, ease of use, effect of source specification and to investigate their sensitivity to input data and mesh resolution, and in particular the effect of different model formulations and assumptions followed by the models.

The possibility of identifying emission sources in the near and far field is investigated, by modelling dispersion backwards in time, in particular the discrimination of multiple sources from receptor data is discussed. The effect of meteorological data resolution on the output of LPD models was evaluated and the most suitable methodology for better source definition was determined for different modelling scales, ranging from the intercontinental transport of airborne pollutants to simulating pollution episodes caused by local sources.

The project ATTMA

In the framework of the cross-border EU Interreg IIIA activity, the joint Anglo-French project ATTMA was commissioned to study Aerosol Transport in the atmosphere of the Cross-Channel, or "Trans-Manche" region of Nord-Pas-de-Calais (France) and Kent (Great Britain).

The air quality of the region is dominated by the industrial area of Dunkerque, in addition to transportation sources linked to cross-channel traffic in Kent and Calais and Channel shipping.

The objective of ATTMA project was to develop and promote tools, which improve the assessment of Air Quality in the Euroregion. The ATTMA project studied suspended air particles, drawing special attention to the proportion of fines, i.e. PM2.5. ATTMA is linked to the French-Flemish EXPER/PF database which collected Particulate Matter (PM) measurements from various local air quality networks. This information was complemented by PM maps obtained from remote sensing data (SeaWiFS and MERIS satellites). Furthermore the ATTMA project researches the particulate matter transport within the Trans-Manche atmosphere whilst broadening the use of remote sensing data and atmospheric dispersion modelling methods (aims of this research) in order to promote a closest collaboration and to extend the use of the project for the purpose of health impact assessment.

The partners involved in this project cover a large part of the eligible territory in the fields of environmental research or Air Quality Monitoring (Table 1).

Project Partners	**Partner Activities**
ADRINORD (Association for the Development of Research and Innovation, Nord-Pasde- Calais)	Remote sensing data and in situ measurements
Autorité de Gestion (AG) Conseil Régional de Haute Normandie Cellule INTERREG IIIA	Local Authority
ATMO Nord-Pas de Calais	*(free contributor)*
APPA Association for Prevention of Atmospheric Pollution	Remote sensing data and in situ measurements
AREMA Lille Métropole	Air Quality Network for the zone of Lille
The University of Greenwich (a) School of Computing and Mathematical Sciences (CMS)	Atmospheric Dispersion Modelling(This Research)
The University of Greenwich (b) Natural Resources Institute	Source Apportionment of Characterisation of Airborne Particles
Kent County Council Environmental Management	Local Authority

Table 0.1 ATTMA project partners

PROJECT PARTNER	PARTNER ACTIVITIES
ADRINORD Association for the Development of Research and Innovation, Nord-Pasde- Calais	Project leader
APPA Association for Prevention of Atmospheric Pollution	Communication (France)
AREMA Lille Métropole Air Quality Network for the zone of Lille	Measurements and database Feeding
HYGEOS Hydrogeology, Space Observation, Environment	Construction of the database
OPAL'AIR Air Quality Network for the zones of Boulogne, Calais, Dunkerque and Saint-Omer	Measurements & database Feeding
VLIZ Vlaamse voor de Zee vzw Coordination & information of maritim research in Flanders	Communication (Flanders)
V.M.M. Vlaamse Milieumaatschappij Environmental flemmish Agency	Measurements & database Feeding

Table 0.2 EXPER/PF project partners (source: Schadkowski C, 2004)

Contents

1. Introduction

Atmospheric pollution is a broad term applied to any chemical, physical (particulate matter), or biological substances that in certain levels or concentrations can modify the natural characteristics of the atmosphere in a way that it may impair the health of plants and animals (including humans) reduce visibility or affect the climate.

Meteorological science is the study of the atmosphere, the complex dynamic natural system that is essential to support life on planet earth. Poor air quality due to international airborne pollutants is responsible for various environmental and human health problems. Although meteorology is an old and well established science, air quality science has a much shorter history and although problems of poor air quality were experienced before the twentieth century, they were usually treated as regulatory issues rather than as a separate science (*Boubel et al. 1994*).

The first attempt of controlling atmospheric pollution occurred in 1361, when King Edward I of England banned by proclamation, the combustion of sea-coal in London, after its smoke had become an issue of health and comfort (*Morgan et al, 1984*). The Public Health Act of 1848 began the process of waste regulation in Great Britain, identifying the need for clean air, clean water, healthy and appropriate nourishment and accommodation as being essential for good health (*Alderslade, 1998*).

Recently, monitoring of air quality has become one of the most active areas of research within the European Union, due to the fact that poor air quality still affects human health in most of the countries of the European region, despite the effort to reduce the concentrations of many pollutants (*World Health Organisation Regional Office of Europe, 2006*). A variety of outdoor and indoor airborne pollutant sources contribute to health risks, and the harmful properties of many common pollutants are still under intensive research.

The British Parliament's Environment Act 1995 created a number of new agencies and set new standards for environmental management. It set up:

- The Environment Agency
- The National Park authorities
- The Scottish Environment Protection Agency (SEPA)

The Environment Act 1995 and subsequent regulations provided a statutory duty for Local Authorities to review air quality in relation to specified pollutants. The examined pollutants, known to have an effect on health, to some degree or other, were (*Environment Act, 1995*):

- 1,3-Butadiene
- Benzene
- Carbon Monoxide
- Nitrogen Dioxide
- Ozone
- Particulate Matter
- Sulphur Dioxide
- Lead

The European Council aimed to develop a comprehensive strategy through the setting of long-term air quality objectives. A series of Directives were introduced to regulate levels of certain pollutants and to monitor their concentrations in the air. The EU-Framework Directive on Air Quality was developed on the 27th of September 1996 and established a list of 13 pollutants for which requirements should be described in subsequent Daughter Directives. The first Daughter Directive, namely 1999/30/EC, gave air quality norms (limit values and alarm thresholds) for sulphur dioxide (SO2), nitric or nitrogen oxides (NOx), PM10 and lead. The second directive, namely 2000/69/EC gave limit values for carbon monoxide (CO) and benzene. The third Directive (2002/03/EC) proposed target values and thresholds for ozone. No legal limit values were proposed by the EU-Commission for ozone, as it does travel over borders, and also it is a secondary pollutant, whose production is highly influenced by meteorological parameters (*INTERREG III Project EXPER/PF, 2005*).

Different methods exist for evaluating air quality. Such methods include the use of direct observation and measurement, mathematical models, remote sensing and air quality indicators, such as those derived from effects on the environment or human health. The most universally accepted methods are through measurements of the concentrations of specific atmospheric substances near to ground level and through the use of a variety of mathematical models to predict the transport and dispersion of airborne pollutants in the lower atmosphere. Other methods include the use of remote sensing tools, for example by the analysis of satellite data.

This research is concerned with the application of mathematical models to numerically simulate the dispersion of airborne pollutants in the lower atmosphere. Data from air quality monitoring stations and remote sensing data were used to initiate the models (input data) or to validate the output results.

It is often required to identify the source of pollution which may lie outside the limits of jurisdiction of any local authority. For this reason inverse (or receptor) modelling can be applied to draw source-receptor probabilities (*Flesch et al., 1995*) (*Seibert et al., 2004*). It is one of the objectives of this work to investigate the applicability of inverse modelling techniques in air quality assessment.

As many important problems related to environmental and health impact assessments and other quantitative policy studies involve issues of science or engineering that are not well defined, such as meteorological forecasting, it is of great importance to adequately characterise and deal with uncertainties such as as those associated with air quality modelling techniques. It is one of the primary objectives of this work to investigate the accuracy and sensitivity of the available atmospheric dispersion modelling tools.

1.1 Lagrangian Particle Dispersion Models

The simulation of the transport and dispersion of pollutants from their sources and within a moving fluid, such as the Earth's atmosphere may be performed by two approaches: the Eulerian or the Lagrangian.

The Eulerian approach calculates the pollutant concentration changes over a fixed gridded region (domain) based on numerical calculations of the basic conservation equations (i.e. mass, momentum, energy) and with the assistance of appropriate initial and boundary conditions. Although the Eulerian modelling permits simple treatment of nonlinear processes arising from interactions of pollutants from multiple and diverse sources (i.e. photochemistry), the approach is often criticised for its limitations in the spatial resolution of the processes leading to high computational complexity and the appearance of artificial diffusion (*Nielinger et al, 2004*; *A. Stohl, 2005*).

Lagrangian Particle Dispersion (LPD) models compute trajectories of a large number of notional particles and can be used to numerically simulate the dispersion of a passive tracer in the lower atmosphere. Each Lagrangian particle carries a certain amount of gaseous or aerosol mass. The particle transport is then determined by average wind velocity components and turbulence conditions, modelled in most Lagrangian particle models as a Markov process using linear Langevin stochastic differential equations (*Gifford, 1982; Sawford, 1984*). The concentration is usually calculated by counting particles or time-intervals of particles within the grid volumes. Although artificial numerical diffusion does not occur with the Lagrangian method, a vast number of particle-trajectories have to be calculated (since statistical accuracy is important), which may lead to computing resource-consuming simulations, especially, when a high resolution domain is required (*Nielinger J., 2004*). Lagrangian Particle Dispersion models are described in further detail in Chapter 3.

Fisher et al (2002) have shown the advantages of applying two or more dispersion models for assessing the air quality impact of atmospheric pollutant sources, each of which is reported to perform better than the other in certain circumstances. This research uses and compares two widely used particle dispersion models: the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model and the model FLEXPART. Both models possess the forward tracking and inverse (or receptor-based) modes. Both models in this study use meteorological datasets derived from the MM5 mesoscale model or datasets from the ECMWF MARS archive which are also used as initial and boundary conditions for the MM5 model. The two models make use of the available meteorological data in different ways and it is important to understand the assumptions made by each model and the implications for the accuracy in defining trajectory paths and source regions. These differences will be explained in Chapters 4 and 5.

1.2 Objectives of the Research

This thesis is submitted as part of the research: "Aerosol Transport in the Trans-Manche atmosphere - Lagrangian particle dispersion and trajectory modelling" that investigates the air pollution dispersion across the English Channel, in collaboration with local authorities and other universities in Southern England and France. The specific aims of the research are:

- To determine, through modelling, pollutant dispersion patterns within the Trans-Manche region and identify sources responsible for episodes detected, using ground monitoring data from British and French networks and with the assistance of satellite images.
- To develop and introduce a dispersion modelling methodology that can be applied by the for air quality assessment within the Trans-Manche region.
- To investigate the accuracy and the limitations of dispersion modelling tools and the spectrum in which they can be applied to assist the air quality assessment
- To evaluate the effect of meteorological data resolution on the output of LPD models
- To determine the most suitable methodology for better source description for different modelling scales, ranging from global to local domain sizes.
- To improve the Lagrangian dispersion modelling tools.

1.3 Outline of present contribution

This research made a number of significant and original contributions in the area of Lagrangian Particle Modelling. Contributions described in this dissertation can be summarised as follows:

- Two widely used Lagrangian Particle Dispersion models, namely FLEXPART and HYSPLIT were compared and their differences were explored.
- The major recent pollution episodes that affected the area of Trans-Manche were investigated and their sources where identified through receptor modelling.
- As part of this research various input/output facilities for the community based Lagrangian Particle dispersion model FLEXPART were developed, and became then available to the international FLEXPART community.
- The MM5 version of FLEXPART (version 3.1) was maintained, and features from the ECMWF reference version (version 6.2) were introduced.
- Performance was demonstrated with real world examples of the sensitivity of the LPD models to grid resolution, input data and configuration settings.

1.4 Airborne pollution

Since this research is concerned with air quality modelling it is important to introduce certain useful definitions regarding the atmosphere and air quality. First, the basic characteristics of the Earth's atmosphere are described as regards to air flow dynamics and atmospheric pollutants. The section continues with an overview of methods for air quality assessments, namely ground based sampling, remote sensing and air quality modelling.

1.4.1 The Earth's Atmosphere

Earth's atmosphere is a complex, dynamic system that is essential to support life on the planet. Discussion of the vertical profile of Earth's atmosphere is needed to understand the dynamics of airborne pollutant dispersion in the atmosphere. The vertical profile of atmosphere may be divided into several distinct layers namely:

- The **Troposphere** made up of the layer called **Atmospheric Boundary Layer (ABL)** or the **Planetary Boundary Layer** (PBL) and the **free troposphere**;
- The **Stratosphere** and the Ozone Layer;
- The **Mesosphere** and **Ionosphere**.

Many atmospheric dispersion models are referred to as boundary layer models because they can model air pollutant dispersion mostly within the ABL. This research also concentrates on the Atmospheric Boundary Layer (ABL).

The Troposphere: The troposphere is lowest layer of the atmosphere and where most weather phenomena take place; it is the region of rising and falling parcels of air. It counts for about 80 percent of the mass of the overall atmosphere and ranges in thickness from 8 km at the poles to 16 km over the equator. The air pressure at the top of the troposphere is only 10% of that at sea level (0.1 atmospheres).

The troposphere can be divided into two parts: The lowest part namely the **Atmospheric Boundary layer (ABL)** or **Planetary Boundary Layer (PBL)** extending upward from the surface to a height that ranges from 100 m to 3000 m, and above it, the **free atmosphere**. The PBL is of the most important layer regarding the emission, transport and dispersion of airborne pollutants. In the PBL layer physical quantities such as flow velocity, temperature, moisture etc., display rapid fluctuations (turbulence) and vertical mixing is strong. Therefore, in any weather prediction model, the PBL must be parameterised as a mechanism for turbulence (Stull, 1988). The air temperature of the PBL decreases with increasing altitude until it reaches the **inversion layer** that caps the atmospheric boundary layer. In the inversion layer the temperature increases with increasing altitude.

Above the PBL is the free atmosphere where the wind is approximately gaiostrophic (parallel to the isobars) while within the PBL the wind is affected by surface drag and turns across the isobars. The free atmosphere is usually non turbulent, or only intermittently turbulent.

The Stratosphere and Ozone Layer: The next layer above the troposphere is the stratosphere, where air flow is mostly horizontal. It is situated between about 10 km and 50 km altitude above the surface at moderate latitudes, while at the poles it starts at about 8 km altitude. This layer plus the troposphere make up 99% of the total mass of the atmosphere. The ozone layer in the upper stratosphere has a high concentration of ozone. This layer is primarily responsible for absorbing the ultraviolet radiation from the Sun. There is considerable recent concern that specific compounds released by natural processes or human activities may be depleting the ozone layer, with serious future consequences for life on the Earth.

The Mesosphere and Ionosphere: Above the stratosphere is the mesosphere and above that is the ionosphere (or thermosphere), where many atoms are ionised (have gained or lost electrons so they have a net electrical charge).

1.4.2 Airborne pollutants and their effect on human health

1.4.2.1 Overview

Atmospheric pollution, is a broad term applied to any chemical, physical (particulate matter), or biological substances that in certain levels or concentrations can modify the natural characteristics of the atmosphere in a way which may impair the health of plants and animals (including humans), or reduce visibility.

Air pollution caused by natural processes has occurred on Earth since the planet's formation: Phenomena such as forest fires, volcanic eruptions, desert dust storms, meteorite impacts, and high winds are all natural sources of air pollution. Anthropogenic problems of poor air quality were not observed before the 16th century. In the nineteenth and early twentieth centuries, the majority of problems related to poor air quality were caused by chimney and smoke stack emissions of coal combustion products and the chemical industry (Y. Ji and S. Plainiotis, 2006). Since the 1970s, further environmental problems are increasingly recognised to be related to atmospheric pollution, such as regional acid deposition, global ozone reduction, Antarctic ozone depletion, and global climate change (Table 1.1).

The **EU Framework Directive 96/62/EC** on ambient air quality assessment and management was adopted by the Environment Council in 1996. This Directive revised the previously existing legislation and the introduced new air quality standards for previously unregulated air pollutants, setting the timetable for the development of daughter directives on a range of pollutants.

The list of atmospheric pollutants to be considered included pollutants governed by already existing ambient air quality objectives:

- sulphur dioxide,
- nitrogen dioxide,
- particulate matter,
- lead and
- ozone

and introduced new pollutants:

- benzene,
- carbon monoxide,
- polycyclic aromatic hydrocarbons and
- cadmium, arsenic, nickel and mercury.

The European Council **Decision 97/101/EC** established a Community-wide procedure for the exchange of information and data on ambient air quality in the European Community. The decision introduced a collaboration and mutual exchange of air quality information, as regards to the national air quality monitoring networks and stations of the member States. This information exchange relates to the pollutants that were listed in Annex I of Directive 96/62/EC.

Commission **Decision 2001/752/EC** amended the Annexes of the Decision 97/101/EC. A Guidance report on the Annexes to Decision 97/101/EC on Exchange of Information as revised by Decision 2001/752/EC is now available.

Pollutants can travel long distances, chemically reacting in the atmosphere to produce other pollutants, such as acid rain or ozone. Pollutants can be classified as either primary or secondary. Primary pollutants are directly produced by a process, such as ash from anthropogenic combustion or volcanic eruption. Secondary pollutants are not directly emitted, but formed from the reaction or interaction of primary pollutants and compounds. One of the most important secondary pollutants is ozone, the product of the reaction between many pollutants that make up photochemical smog. Pollutants are physically removed from the atmosphere by dry and wet deposition. Dry deposition is a removal process that occurs when a gas or particle collides and sticks to a surface such as the Earth's surface terrain or vegetation. Wet deposition of a pollutant occurs by dissolution in precipitation (rainfall) that falls to the ground.

The health effects associated with air pollution arise through a series of different processes such as physical, chemical, physiological and behavioural etc. and various methods exist for assessing the health impact of airborne pollutants (Y. Ji and S. Plainiotis, 2006). The following paragraphs provide an overview of the main health issues associated with the major airborne pollutants.

1.4.2.1 Sulphur dioxide (SO_2)

Sulphur dioxide in the atmosphere is a gaseous pollutant, which is usually encountered at high concentrations around industrial areas and arises from both natural and human activities. Natural processes which release sulphur compounds include decomposition and combustion of organic matter; spray from the sea; and volcanic eruptions. The main human activities producing sulphur dioxide are the smelting of mineral ores containing sulphur and the combustion of fossil combustible (coal, oil) by industry and domestic heating. In water, it dissolves to form sulphuric acid, a corrosive substance that damages materials and the tissue of plants and animals. It is responsible for respiratory damage in the case of high concentrations (Table 1.2). Ambient concentrations have decreased in recent years as the same time as improvements in industrial processes have occurred.

In the Trans-Manche region, there is particular concern in coastal and particularly port areas, where diesel oil used in shipping is a major SO_2 contributor.

HOURLY LIMIT VALUE for the PROTECTION of HUMAN HEALTH	350 μg/m³ not to be exceeded more than 24 times a calendar year
DAILY LIMIT VALUE for the PROTECTION of HUMAN HEALTH	125 μg/m³ not to be exceeded more than 24 times a calendar year
ALERT THRESHOLD	500 μg/m³ over 3 consecutive hours

Table 1.2 Limit values for the protection of health *for SO2 coming into force on 1/1/2005 and alert threshold for SO_2 in 2001 (1999/30/EC) (Interreg III Project EXPER/PF, 2005)*

1.4.2.2 Airborne Particulate Matter

This refers not to gaseous pollutants, but to solid or liquid particles which may be suspended in the air. Airborne particulate matter is a mixture of solid and liquid materials of various sizes, ranging from a few nanometres in diameter to around 100 micrometres (100 μm.). Particulate matter consists of complex chemical, physical and biological substances, differing from place to place and from time to time. The ambient atmosphere contains non-biological material (such as dust, smoke, sea salt etc) and a mass of biological material, in the form of bacteria, fungal spores and pollens, many of which are known to cause infection and allergic disease.

Although the precise mechanism by which particles damage health remains unclear, a large number of epidemiological studies have shown relationships between various indices of ill health and measurements of particle concentrations. Particulate matter can be associated with a range of effects on health including effects on the respiratory and cardiovascular systems, asthma and mortality (Defra, 2002).

Particles in the atmosphere may be a primary pollutant, such as smoke particles, arising from combustion sources (including road traffic), or a secondary pollutant formed from the chemical reaction of gaseous pollutants, (mainly sulphate and nitrate formed by chemical reactions in the atmosphere), or exist in the form of coarse particles, suspended soils and dusts, sea salt, biological particles, tyre rubber and particles from construction work.

Airborne particles can be classified by their average size. The term PM10 refers to the concentration of particles that have diameter less than or equal to around 10μm (table 1.3). Similarly, the term PM2.5 describes the concentration of particles with diameter less than or equal to 2.5μm (table 1.4). Large particles usually settle out of the air (dry deposition), while smaller particles may remain suspended for days or months. Wet deposition (rainfall) is another important mechanism for removing particles from the air.

The size of a particle also determines its potential impact on human health. Larger particles are usually trapped in the nose and throat and swallowed, while smaller particles may reach the lungs and cause irritation there.

There is strong evidence (*Monn, and Becker, 1999*) (*Samet, et al, 2000*) to conclude that fine particles – usually measured as PM2.5 (table 1.4) in health effects studies – are more hazardous than larger ones. Recent toxicological studies show that particles originating from internal combustion engines, coal burning, residual oil combustion and wood burning have strong inflammatory potential (*EXPER/PF, 2005*).

Legislation	24-hourly limit values and	Annual limit values and objectives	Achieve by
EU First Daughter Directive (99/30/EC)	50 μg/m³ with up to 35 exceedences a year	40 μg/m³	1 January 2005
UK Air Quality Strategy (2000)	50 μg/m³ with up to 35 exceedences a year	40 μg/m³	31 December 2004

Table 1.3 EU limit values and the UK Air Quality Strategy objectives for particulate matter (measured as PM10). (Adapted from AQEG, 2005)

DAILY LIMIT VALUE	65 μg/m³
ANNUAL LIMIT VALUE	15 μg/m³

Table 1.4 Limit values for PM2.5 from the US Environmental Protection Agency (EPA) (Adapted from EXPER/PF, 2005)

1.4.2.3 Nitrogen oxides

The term nitrogen oxides is used to refer to any of these oxides (oxygen compounds) of nitrogen, or to a mixture of them. The nitrogen oxides are generated by all combustion sources (industrial plus domestic) especially by motor vehicles, as a result of the reaction between nitrogen and oxygen at high temperature (thermal NOx) or due to Nitrogen in the fuel (fuel NOx).

Once emitted, nitrogen oxide is oxidised by atmospheric oxygen to form nitrogen dioxide, especially in warm sunny conditions. Oxides of nitrogen may remain in the atmosphere for several days and may react to generate nitric acid, nitrates and nitrites as particles. Nitrogen oxides have an important role in the chemical reactions which generate photochemical smog. At high concentrations nitrogen dioxide can provoke respiratory damage.

1.4.2.4 Carbon monoxide

Carbon monoxide is an odourless, colourless gas produced by incomplete oxidation (burning). Its principal source is from the engine of vehicles. Carbon monoxide is also produced by natural processes, such as by the oxidation in the oceans and air of methane produced from organic decomposition. As a pollutant it may remain in the atmosphere for long periods (1 month or more) and is generally found in high concentrations near important traffic roads or in places with bad dispersion (canyon streets or indoors). When high doses are inhaled, CO binds to the oxygen-carrying site on the blood's haemoglobin, which reduces oxygen transport in the body, leading even to death. At high concentrations it causes headaches, reduced ability to focus and concentrate, and nausea.

1.4.2.5 Ozone (O_3)

Near the ground, ozone is a colourless, airborne pollutant. Ozone is often classified as a secondary pollutant as it is not directly emitted from human activity or natural processes, but is formed by a photochemical reaction between nitrogen oxides under the influence of sunlight which is more effective under high temperatures. Ozone is also usually produced in office environments from photocopying machines. Ozone provokes respiratory damage and ocular problems. At high concentrations, it can provoke an asthmatic crisis in sensitive persons. Also it is strongly oxidising and can irritate the eyes and damage vegetation. (King's College - London Air Quality Network: http://www.londonair.org.uk)

Once formed, it can remain in the atmosphere for several days and may be transported over long distances. Studies have shown that European ozone levels have increased rapidly since 1940 and monitoring data from rural sites in the UK suggest that there was a small annual increase during the 1990s (King's College - London Air Quality Network: http://www.londonair.org.uk).

The European Directive 92/72/EEC describes the threshold values for the ozone concentrations in ambient air that apply for the protection of human health and for the protection of the vegetation (table 1.5).

The formation of ozone in the 'stratosphere' is due to a different process. There, it is not considered as a pollutant, but as an important element in absorbing harmful ultraviolet radiation and preventing radiation from reaching the Earth's surface.

INFORMATION THRESHOLD POPULATION	hourly average: 180 µg/m3
ALARM THRESHOLD POPULATION	hourly average: 360 µg/m3
HEALTH POPULATION	8-hours average: 110 µg/m3 (*)
VEGETATION	hourly average : 200 µg/m3, daily average: 65 µg/m3

Table 1.5 Ozone thresholds for the protection of human health and vegetation valid until 8/9/2003 (adapted from Interreg III Exper/PF, 2005)

1.5 Scales of atmospheric motion

1.5.1 Overview

Air pollution dispersion phenomena are strongly influenced by atmospheric processes which cover a wide range of physical scales, from the molecular mechanisms (i.e in water droplet condensation) to the continental size processes involved in the creation of high pressures.

A proper scaling will facilitate the choice of appropriate approximations of the governing modelling techniques. The following paragraphs give an overview of the definitions and descriptions of the existing classification systems used in this thesis.

1.5.2 Ligda's classification

Ligda (1951) classified the atmospheric motions into three scales, based on radar storm observations:

a) **Microscale**: $L < 20$ *km,*

b) **Mesoscale**: 20 *km* $< L < 1000$ *km, and*

c) **Synoptic scale**: $L > 1000$*km,*

where L is the horizontal scale length of the atmospheric motions.

Horizontal Scale	Lifetime	Stull (1988)	Pielke (2002)	Orlanski (1975)	Thunis and Bornstein -1996	Atmospheric Phenomena
10 000km	1 month	Macro	Synoptic Regional	Macro- α	Macro-α	General circulation, long waves
				Macro-β	Macro-β	Synoptic cyclones
2000 km	1 week			Meso-α	Macro-y	Fronts, hurricanes, tropical storms, short cyclone waves, mesoscale convective complexes
200 km	1 day	Meso	Meso	Meso-β	Meso-β	Mesocyclones, mesohighs, supercells, squall lines, inertia-gravity waves, cloud clusters, low-level jets
20 km	1h	Micro	Micro	Meso-γ	Meso-γ	Thunderstorms, cumulonimbi, clear-air turbulence, heat island,
2 km				Micro-α	Meso-δ	Cumulus, tornadoes, microbursts, hydraulic jumps
200 m	30 min			Micro- β	Micro- β	Plumes, wakes, waterspouts, dust devils
20 m	1 m	Micro-δ		Micro-γ	Micro-γ	Turbulence, sound waves
2m	1 sec				Micro-δ	

Table 1.6 Atmospheric scale definitions, where L is horizontal length of the atmospheric motions. (Adapted from Thunis and Borstein 1996)

1.5.3 Orlanski's classification

In 1975, Orlanski proposed a more detailed classification to Ligda's classification and recommended distinguishing 8 scales and subscales: microscale (subscales: *-α, -β, -γ*), mesoscale (subscales: *-α, -β, -γ*) and macroscale (subscales: *-α, -β*) (Orlanski, I., 1975):

a) **Microscale**: *Lh < 20 km*

Air flow patterns are very complex at this scale, as they are dominated by hydrodynamic characteristics, such as the terrain roughness, building characteristics, or flow channelling. Due to the complex nature of those effects, micro-scale dispersion phenomena are usually described with robust local models in the case of practical applications, such as street canyon models (Moussiopoulos, *1996*), (Vardoulakis *et al., 2003*). The micro-scale is further divided by Orlansky (1975) into the following sub-scales:

- micro- α (2 km > L > 200 m),
- micro- β (200 m > L > 20 m),
- micro- γ (L < 20 m) subscale

b) **Mesoscale:** *20 km < L < 22000 km,*

Mesoscale atmospheric phenomena affect primarily local-to-regional scale dispersion phenomena, for which urban studies are the most important examples. On this scale flow patterns are influenced not only by hydrodynamic effects but also by inhomogeneities of the energy balance, mainly caused by the spatial variation (e.g.

land use) and also by topographic characteristics. Thermal effects are also an important aspect especially in times of weak synoptic forcing (Moussiopoulos, *1996*).

Therefore, mesoscale models should be capable of simulating local circulation systems, such as sea and land breezes. Furthermore, the description of urban-level phenomena that are confined within the mesoscale requires the utilisation of fairly complex modelling. Mesoscale is further divided into three subscales:

3. meso-α (2000 km > L > 200 km),

3. meso-β (200 km > L > 20 km),

3. meso-γ(20 km > L > 2 km),

c) **Macroscale** *(characteristic lengths exceeding 1,000 km);*

Global and most regional-to-continental scale dispersion phenomena take place within the macro-scale of atmospheric processes. At this scale, the atmospheric flow patterns are mainly influenced by synoptic phenomena, such as the geographical distribution of pressure system, usually caused by large-scale inhomogeneties of the surface energy balance. Hence the hydrostatic approximation can be considered as valid when modelling in this scale. The macroscale can be further divided into:

- macro-α (L > 10,000 km) and
- macro-β (10,000 km > L > 2000 km) scale.

1.5.4 Emanuel and Raymond classification

Emanuel and Raymond (1984) defined the following different scales of atmospheric phenomena:

(a) **Synoptic scale** - for motions which are quasi-geostrophic and hydrostatic,

(b) **Mesoscale** – for motions which are *non*-quasi-geostrophic and hydrostatic, and

(c) **Microscale** – for motions which are non-geostrophic, non-hydrostatic, and turbulent.

Therefore, the **mesoscale** may be defined as the scale, which is large enough in the horizontal dimension to be considered hydrostatic, but not quasi-geostrophical.

1.5.5 Other classification systems

Recent texts have proposed alternative scaling for atmospheric phenomena:

Pielke in 1984, defined mesoscale phenomena as those having a horizontal length scale large enough to be hydrostatic, but small enough to be highly influenced by the Coriolis force (small enough in comparison to the advective and pressure gradient forces). His definition of mesoscale, therefore, confined with the definition of Orlanski's meso-□scale (Table 1.6). Pielke (1984) splits the meso-α and macro scales of Orlanski into regional and synoptic scales and defines the vertical bounds of mesoscale phenomena as extending "from tens of metres to the depth of the troposphere".

Stull (1988), basing his classification on Orlanski, added the micro-□ extended from 2 m to 2 mm and defined the lower limit of the mesoscale at 3 km.

Thunis and Bornstein (1996) took a different approach in their classification which is based on some assumptions, such as hydrostatic, convection, advection, approximations of the governing equations, both time, horizontal and vertical scales, to standardise nomenclature for mesoscale concepts and to integrate existing concepts of atmospheric space scales, flow assumptions, governing equations, and resulting motions into a hierarchy useful in the categorization of mesoscale models.

In this research we are not interested in the micro-scale for which the source of pollution regulated by air quality management is readily apparent (road transport, industrial chimneys etc.), nor are we interested in global pollution, since the lifetime of the pollutants regulated by air quality management are generally a few days, although the methods used here can be extended to this using for example meteorological input data of higher resolution. This thesis is focused on methods of identifying sources of pollutants which have travelled for a short period of usually two or more days, for which trajectory methods are appropriate (mesoscale). Over this scale, dispersion processes are more important than details of the transport or advection phenomena.

1.6 Methods for assessing air quality

Developing plans for air quality management is in most cases performed within the following four phases: *monitoring, modelling, policy planning* and finally *by policy execution.* Air quality monitoring and air quality dispersion modelling are very important phases for understanding the nature of air pollution, which in turn is crucial for developing effective plans for air quality management and protecting the environment and public health. Monitoring is usually achieved by two methods: surface (receptor) observations and remote sensing.

1.6.1 Air Quality Monitoring

Data collected as part of the air quality assessment are used for evaluating current pollution concentrations, trends, and compliance with air quality standards. Air quality assessment may also include an initial identification of pollution sources through careful analysis of the monitoring data.

Monitoring can be performed with the assistance of mobile stations and fixed air quality stations that can be divided into kerbside, roadside, urban centre, urban background, urban industrial, suburban, rural and remote according to the measurement location (Table 1.7). More information about monitoring methods adopted in this research is given in Chapter 3.

Kerbside	Sites with sample inlets within one metre of the edge of a busy road.
Roadside	Sites with sample inlets between one metre of the edge of a busy road and the back of the pavement (usually five metres from the roadside).
Urban centre	Sites away from roads in city and town centres (for example, in pedestrian precincts and shopping areas).
Urban background	Sites in urban locations (for example parks and urban residential areas) away from specific emission sources. These locations broadly represent city-wide background concentrations.
Urban industrial	Sites where industrial emissions can make a significant contribution to measured pollution concentrations.
Suburban	Sites typical of residential areas on the outskirts of a town or city.
Rural	Sites in the open country away from roads and industrial and residential areas.
Remote	Sites in the open country in isolated rural areas that only have regional background pollution concentrations for most of the time.

Table 1.7 Definitions of types of monitoring site (Source: Air Quality Expert Group, 2005)

1.6.2 Air quality monitoring using remote sensing technology

Data requirements to support air quality management decisions and policies need to be expanded to large spatial domains in order to accommodate policies, which frequently cross geopolitical boundaries; from microscale and mesoscale to the macroscale. On the other hand, as air pollution continues to cross geopolitical boundaries, the need for more frequent assessments to help combat global air pollution will increase. It has been found that remote sensing air monitoring equipment has the capability to provide information on critical environmental variables that involve broad spatial scales and require data that can provide synoptic information. In addition, large ground level monitoring networks are impractical to implement on a global basis.

1.6.2.1 The physics of remote sensing technology

A fraction of the solar incident light is reflected by the Earth towards space. This radiation, in one given direction, can be observed by satellite sensors (radiance in $W/m^2/sr$) reflected by the atmosphere-surface system in the spectral domain from the blue (412 nm) to the near infrared (865 nm) (INTERREG III Project ATTMA, 2005).

From space, one can observe the aerosols particularly during strong dust episodes, such as those originating from Saharan dust storms across the Mediterranean Sea and over the Atlantic Ocean. However aerosols from local air pollution sources can also be observed as is illustrated in Figure 1.4.

one in the infra red at 865 nm). Over land the brightness of Earth's surfaces makes in most cases the aerosol contribution vanish. Exceptions include surfaces covered with strong vegetation where the chlorophyll (photosynthesis) strongly absorbs light in the blue (443 nm) and in the red (670 nm) (INTERREG III Project EXPER/PF, 2005).

Remote sensing projects, important for air quality modelling include: MODIS, MISR, AVHRR, DMSP, GOES and Total Ozone Mapping Spectrometer *(TOMS)* (Herman *et al., 1997*) and those most closely related to this research are described below.

The TOMS Aerosol Index (AI) is an indicator of smoke and dust absorption. It is calculated on the basis of multispectral measurements in the ultraviolet (UV) wavelengths derived from the TOMS/OMI instrument, which allows the detection of UV absorbing aerosols over both land and sea surfaces. The index is unitless and is scaled by 10. For better visualisation only data greater or equal to 7 are mapped. The AI data from TOMS and OMI instruments are available free from the NASA Goddard Earth Sciences Data Information Services Center (GES DISC) (http://acdisc.gsfc.nasa.gov).

The absorbing Aerosol Index is a qualitative parameter. However, it is an excellent tool in classifying UV absorbing and non-absorbing aerosols (Ahmad *et al.*, 2006). A major limitation of AI data is the fact that the vertical profile of the pollutants cannot be determined from overhead satellite measurements, as the index is cumulative of all levels from near the ground to the observation point (total column). Other limitations include the low resolution (50 km) of the currently available AI data from TOMS and OMI (Torres *et al.,* 1998) and probable verification problems near the ground (below 1.5 m) or under cloudy conditions, where sensitivity is reduced (Herman et al, 1996).

Data from the TOMS and OMI instruments were used in this research for the qualitative determination of the position of dust plumes originating from desert dust storms. Output from the LPD models was superimposed on AI data and the relative position of the dust plumes were compared (see Chapter 5).

1.6.2.3 The Moderate Resolution Imaging Spectro-radiometer (MODIS) Sensor

The Moderate Resolution Imaging Spectroradiometer (MODIS) of the USA's NASA is used for studies of various pollutants (Esaias *et al,* 1998).

MODIS instrument consists of a scan mirror collecting optics, and individual detector elements. Imagery is provided in each of 36 different spectral bands ranging from the visible to the infrared portions of the electromagnetic spectrum, from 0.4 to 14.54 μm. The spectral bands have a spatial ground-level resolution of 250 m to 500 m, (or 1 km at nadir), signal-to-noise ratio greater than 500 at 1-km resolution, and absolute irradiance accuracy of +/- 5% from 0.4 to 3 micrometer and 1% thermal infrared (3 to 15 micrometer).

MODIS provides daytime reflection and day/night thermal emission imaging at any point on the Earth at least every 2 days. The MODIS instrument flies on the EOS Terra (formally EOS AM-1) and EOS Aqua (formally EOS PM-1) series of spacecraft (Esaias *et al.,* 1998).

1.6.2.4 The MEdium Resolution Imaging Spectrometer Instrument (MERIS)

MERIS of the European Space Agency (ESA) is an imaging spectrometer that flies on the ESA's ENVISAT satellite and measures the solar radiation reflected by the Earth, at a ground spatial resolution of 300m, in 15 spectral bands, programmable in width and position, in the visible and near infra-red. MERIS allows global coverage of the Earth in 3 days. MERIS is also capable of retrieving cloud top height, water vapour total column, and aerosol load over land.

1.6.2.5 The Sea-viewing Wide Field-of-view Sensor (SeaWiFS)

SeaWiFS is on a sun-synchronous (overpass time is around noon) ocean colour sensor on a polar orbit. It provides information at a ground spatial resolution (horizontal) of 1 km by 1 km actual surface, and 24 hours temporal (rarely 12 hours) (EXPER/PF, 2005). It has been operational since 1998 and data are provided by NASA free of charge for non-commercial uses.

The initial purpose of the Sea-viewing Wide Field-of-view Sensor (SeaWiFS) Project was to monitor subtle changes in Earth's ocean colour, signifying various types and quantities of marine phytoplankton (microscopic marine plants). The concentration of phytoplankton can be derived from satellite observation and quantification of ocean colour. This is due to the fact that the colour in most oceans in the visible light region, (wavelengths of 400-700 nm) varies with the concentration of chlorophyll and other plant pigments present in the water (Hooker, 1992). The aerosols are remotely sensed with SeaWiFS over water using spectral bands 6 and 8. (EXPER/PF, 2005).

1.6.2.6 Advanced Very High Resolution Radiometer (AVHRR)

The Advanced Very High Resolution Radiometer (AVHRR) sensor of the National Oceanic and Atmospheric Administration (NOAA) satellite series is used for a wide range of applications in polar and climate research. AVHRR provides four- to six-band multi-spectral data from the NOAA polar-orbiting satellite series and was originally designed for use as an imaging radiometer for meteorological purposes, rather than for quantitative radiometric sensing. However, as new applications evolved, quantitative radiometric data became necessary. There is fairly continuous global coverage since June 1979, with morning and afternoon acquisitions available. The spatial ground level resolution is 1.1 kilometre at nadir (NOAA, 1985). AVHRR satellite data was used in this research for validations with output from LPD model HYSPLIT.

Since it is not possible to cover all aspects of exposure assessment for an airborne pollutant with remote sensing or other monitoring methods, air quality modelling has become an essential research tool. This is covered in the next following sections.

1.6.3 Air quality modelling

Air quality models are representations of the atmospheric processes responsible for air pollution, such as the ozone formation. Two distinctly different methods have been developed for making such (Y. Ji and S. Plainiotis, 2006): mathematical models and physical models. Mathematical (or numerical) air quality models simulate pollutant behaviour by interrelating symbolic descriptions of the important physical and chemical processes occurring in the atmosphere within a computational framework. Physical models simulate atmospheric processes by employing a scaled-down representation of the atmosphere in a laboratory setting. A common example of physical models is wind tunnel simulations using scale-down models of buildings to observe the transport of pollutants in street canyons.

Mathematical models have a number of advantages over physical models such as pollution apportionment capabilities and lower cost. This research is only concerned with mathematical methods of air quality modelling; hence in this study the terms "modelling" and "model" are used to refer to the mathematical modelling methods only.

Mathematical air quality models simulate the atmosphere in varying degree of detail by numerically representing emissions, initial, and boundary concentrations of chemical species; the chemical reactions of the emitted species and of their products; and the local meteorology which may include sunlight, wind, cloud cover and temperature. Hence air quality modelling combines understanding of the atmosphere's chemistry and meteorology with estimates of source emissions to predict possible air pollution concentration and dispersion patterns. Examples of applications of mathematical models to urban air quality can be found in Sokhi and Bartzis (2002). Air quality models are useful tools for:

- understanding the behaviour of various compounds in a complex physical environment
- quantifying ambient concentrations of pollutants or pollutant precursors
- determining optimum locations for air quality monitoring equipment
- determining source contributions to concentration estimates
- developing emission limits for sources
- optimising and evaluating air quality strategies

Mathematical models used in air quality studies fall into two types: empirical/statistical and analytical/deterministic. Empirical/statistical models statistically relate observed air quality data to the accompanying emission patterns. This study is concerned with analytical models, which use analytical expressions to describe the transport and chemical processes involving atmospheric compounds. Analytical air quality models usually consist of a combination of meteorological numerical models with chemical mechanisms and modules derived from atmospheric chemistry research.

The first laboratory attempts to understand the formation of photochemical and urban smog was undertaken in the 1950's and the later availability of computer systems led to the first models simulating atmospheric chemical reactions. In the 1960s and 1970s air-quality models were expanded to two and three dimensions. Such models were used to calculate the emission, transport, chemical modifications and depletion of airborne pollutants. Most models used interpolated fields of meteorological data as inputs, while many modern models today (i.e. FLEXPART, Monte Carlo) use meteorological fields calculated in real time as inputs and many models are able to calculate atmospheric chemistry and dynamical meteorology (i.e. Eulerian Models).

Mathematical air quality modelling methods are described in further detail in Chapter 2.

1.7 Structure of this Thesis

This thesis consists of four main chapters after the Introduction. Chapter 2 offers a review of current methods for Air Quality Modelling. It specifies the location and role of air quality assessment in the causal chain between the emission of pollution and health effects. Chapter 3 describes the methodology used in this research, in the formulations of the Lagrangian dispersion models FLEXPART and HYSPLIT and the input meteorological and air quality data that drive the models. The PSU/NCAR Mesoscale model (MM5) is described, as well as the MARS Archive of the European Centre for Medium-Range Weather Forecasts (ECMWF). Chapters 4 and 5 provide descriptions of case study simulations performed using the above methodology and identifies pollution dispersion patterns within the cross-Channel area of Trans-Manche. Chapter 6 finally describes the issues addressed briefly in the previous sections and presents the conclusions and recommendations emerging from this research. Future work directions, bibliography and auxiliary appendices follow Chapter 6.

Chapter 2

2. Atmospheric Dispersion Modelling Methods

2.1 Overview

This chapter presents a short overview of numerical techniques for modelling the emission, transport, diffusion, transformation, and removal of air pollutants, followed by a summarised description of the most widely used models, their use and purposes.

There is a growing international interest in understanding and predicting the processes by which airborne pollutants are transported in the atmosphere, both for emergency response and planning purposes, and for understanding a range of atmospheric pollution problems. A review of the meteorological and air pollution models available and widely applied within Europe is given by Sokhi et al. (2003).

Moussiopoulos *et al* (1996) distinguished dispersion models on the following grounds:

- on the spatial scale (global; regional-to-continental; local-to-regional; local);
- on the temporal scale (episodic models, (statistical) long-term models);
- on the treatment of the transport equations (Eulerian, Lagrangian models);
- on the treatment of various processes (chemistry, wet and dry deposition) and
- on the complexity of the approach.

Zannetti (1993) distinguished the following model categories of atmospheric dispersion models:

- **Plume-rise models.** Plume-rise models calculate the vertical displacement and general behaviour of the pollutant injected into usually cooler ambient air. Both semi-empirical and advanced plume-rise formulations are available. Both factors (thermal buoyancy and vertical momentum) contribute to increasing the average height of the plume above that of the smokestack.

- **Gaussian models.** The Gaussian plume models are the most commonly used air pollution models. They are based on the assumption that the plume concentration, at each downwind distance, has independent Gaussian distributions both in the horizontal and in the vertical.

- **Semi-empirical models.** This category consists of several types of models which were developed mainly for practical applications. In spite of considerable conceptual differences within the category, all these models are characterised by drastic simplifications and a high degree of empirical parameterisations. Among the members of this model category are box models and various kinds of parametric models. A good review of the application of semi-empirical models in urban street canyons is given by Vardoulakis *et al* (2002).

- **Eulerian models**. The transport of inert or reacting air pollutants may be conveniently simulated by the aid of models which solve numerically the atmospheric advection-diffusion equation, i.e. the equation for conservation of mass of the transported pollutant on a fixed reference frame (the Eulerian approach). Such models are usually embedded in prognostic meteorological models. Advanced Eulerian models include refined sub-models for the description of turbulence which is based on the physics of the flow (e.g. second-order closure models and large-eddy simulation models).

- **Lagrangian Particle Dispersion (LPD) models.** LPD models can be used to numerically simulate the dispersion of a passive tracer in the planetary boundary layer by calculating the Lagrangian trajectories of thousands of notional particles (Zannetti, 1992). LPD models offer particular advantages such as the lack of artificial numerical diffusion, seen on Eulerian models, the ability of source attribution, namely identification of the contribution from particular sources to the total measured or calculated airborne concentration. The methodology of this research is based on LPD models; hence they are further described in chapters 2.3, 3.1 and 3.2.

- **Chemical modules.** Several air quality models include modules to simulate chemical transformation. These schemes have been implemented into dispersion models such as Lagrangian and Euleriar models. In Eulerian models, a three-dimensional grid is superimposed to cover the entire computational domain, and all chemical reactions are simulated in each cell at each time step. In the Lagrangian photochemical models a single cell (or a column of cells or a wall of cells) is advected according to the main wind in a way that allows the injection of the emission encountered along the cell trajectory (Moussiopoulos 1996).

- **Receptor models** also called **inverse models**. In contrast to dispersion models, receptor models start with observed concentrations at a receptor and running with negative time step, seek the apportion of the observed concentrations at a sampling point among several source types. Several existing models (A. Stohl 1996) (R. Draxler, 1997) offer both dispersion (forward) receptor based (backward) modelling capabilities. Receptor models are usually implemented using the Lagrangian method, although recently there have been attempts to develop receptor mode capabilities in Eulerian calculations, using the so-called adjoined approach (Neupauer, 2000).

- **Stochastic models**. Stochastic models are based on statistical or semi-empirical techniques to analyse trends, periodicities, and interrelationships of air quality and atmospheric measurements and to forecast the evolution of pollution episodes. Several techniques are used to achieve this goal, e.g., frequency distribution analysis, time series analysis, Box-Jenkins and other models, spectral analysis, etc. Stochastic models are intrinsically limited because they do not establish cause-effect relationships. However, statistical models are very useful in situations such as real-time short-term forecasting, where the information available from measured trends in concentration is generally more relevant (for immediate forecasting purposes) than that obtained from deterministic analyses.

Table 2.1 summarises, for each scale of dispersion, the separate phenomena and air pollution model categories that have already been used for practical applications (N. Moussiopoulos *et al*, 1996).

2.2 Gaussian Plume and Puff Models

Gaussian plume or puff models have been in use since the 1940s. Their formulation is based on the assumption that dispersion of plumes in the horizontal and vertical direction takes the form of a normal Gaussian curve with the maximum concentration at the centre of the plume (figure 2.1).

Most of the models recommended by the U.S. Environmental Protection Agency (EPA) are Gaussian. Older Gaussian plume models are usually limited to simulating small dispersion distances and small times (up to several hours and up to 50 km). This is because they generally assume constant meteorological conditions, usually producing straight-line trajectories. Modern Gaussian models have been developed in ways that retain the straight-line trajectories of the older Gaussian models, but improve the handling of stability, convection, mixing, and terrain effects (Scire *et al*, 2000), (Cinemorelli *et al.*, 1998).

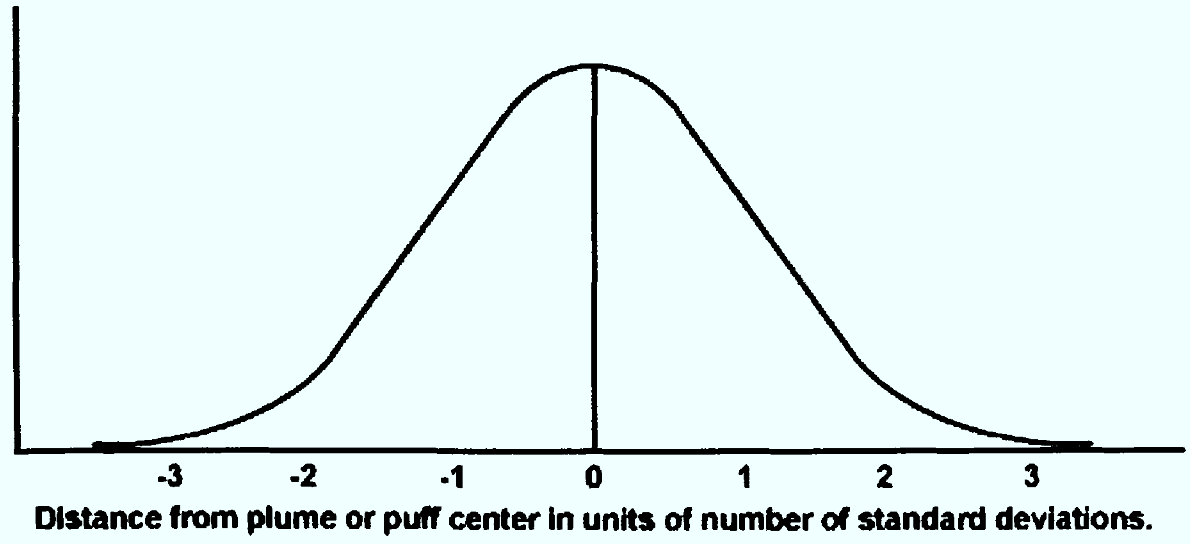

Figure 2.1 Standard deviation in Gaussian plume models Source: The comet program, USA's National Center for Atmospheric Research, USA

The following paragraphs offer a review the Gaussian dispersion models and a brief description of many of the Gaussian models currently in use worldwide.

2.2.1 AMS/EPA Regulatory Model (AERMOD) Modelling System (USA)

The AERMOD modelling system (Cimorelli *et al.*, 1998) is recommended by the U.S.A Environmental Protection Agency as the preferred Gaussian dispersion model for general industrial modelling scenarios (EPA, 1998). It is a steady-state Gaussian plume model designed to handle both flat and complex terrain (Cinemorelli *et al.*, 1998). It uses Gaussian dispersion for stable atmospheric conditions and non-Gaussian dispersion for unstable conditions (high turbulence).

AERMOD consists of two pre-processors AERMET and AERMAP and the dispersion model. AERMET (EPA, 1998) is the meteorological pre-processor and AERMAP (EPA, 1998) is the terrain pre-processor that characterizes the terrain, generates receptor grids and facilitates the generation of hill height scales. AERMOD requires a file of surface boundary layer parameters and a file of profile variables including wind speed, wind direction, and turbulence parameters generated by a meteorological pre-processor.

AERMOD also offers algorithms for plume rise and buoyancy, and the computation of vertical profiles of wind, turbulence and temperature. Sensitivity analysis of AERMET input parameters (Latini G. *et al*, 2001) revealed the importance of these in determining model-predicted maximum concentrations. Algorithms for plume depletion by wet and dry deposition are planned as future additions to the model.

2.2.2 AUSPLUME Model (Australia)

AUSPLUME is a regulatory Gaussian plume dispersion model, developed in around 1986 by the Environmental Protection Authority of Victoria, Australia. AUSPLUME is widely used in Australia to model transport and dispersion within areas of up to few hundreds of square kilometres and makes the following assuptions:

- A simplified meteorology (no horizontal variation) to transport and diffuse pollutants.
- A simplified terrain (flat, or near flat)
- No chemical transformations

2.2.3 The CALPUFF Modelling System (USA)

CALPUFF model has been approved by US Environmental Protection Agency (EPA) as the recommended Gaussian-puff model for assessing long range transport of pollutants and their impacts on Federal Class I areas and on a case-by-case basis for certain near-field applications involving complex meteorological conditions. It is a multi-layer model, multi-species non-steady-state puff dispersion model that includes a diagnostic 3-D meteorological model (CALMET), a puff-based dispersion model (CALPUFF), and a post-processing package (CALPOST). The CALPUFF modelling system is available free from Earth Tech's website: (http://www.src.com/calpuff/calpuff1.htm).

2.2.4 The Advanced Dispersion Modelling System (ADMS) (UK)

ADMS (Carruthers *et al* 1995, CERC 1998) is a state of the art Gaussian-like dispersion model, capable of simulating continuous plumes and short duration puff releases. ADMS is developed by a government and industry consortium in the U.K., led by the Cambridge Environmental Research Consultant (CERC). ADMS also offers algorithms for plume rise and buoyancy, and the computation of vertical profiles of wind, turbulence and temperature. These algorithms are similar to those in AERMOD.

2.2.5 ONM9440 (Austria) (OENORM M 9440, 1996)

ONM9440 is a Gaussian model developed for continuous, buoyant plumes from stationary sources for use in flat terrain areas. It includes plume depletion by dry deposition of solid particulates. Plume rise formulae used in the model are a combination of formulae suggested by Carson and Moses (1969) and Briggs (1975).

2.2.6 The Immission Frequency Distribution Model (IFDM) (Belgium)

IFDM is developed at the Flemish Institute for Technological Research (VITO) for point and area sources dispersing over flat terrain on a local scale. The main concept behind IFDM is the stability classification scheme of Bultynck-Malet (1972) who investigated atmospheric dispersion using the 120 m high meteorological tower of Mol, Belgium. The model calculates plume depletion by dry or wet deposition but is not capable of handling building effects, chemical transformations or complex terrain. IFDM,

2.2.7 HAVAR (Czech Republic)

A Gaussian plume model integrated with a puff model and a hybrid plume-puff model, developed by the Czech Academy of Sciences, is intended for routine or accidental releases of radionuclides from single point sources within nuclear power plants. The model calculates radioactive plume depletion by dry and wet deposition as well as by radioactive decay.

2.2.8 The AERO-POLlution Model (AEROPOL) (Estonia)

AEROPOL is a Gaussian plume model developed at the Tartu Observatory in Estonia for simulating the dispersion of continuous, buoyant plumes from stationary point, line and area sources over flat terrain on a local to regional scale. It calculates plume depletion by wet and/or dry deposition as well as the effects of buildings in the plume path.

2.2.9 STACKS (The Netherlands)

STACKS is a Gaussian plume dispersion model for point and area buoyant plumes to be used for environmental impact studies and evaluation of emission reduction strategies, over flat terrain on a local scale. It includes building effects, NO2 chemistry and plume depletion by deposition.

2.2.10 POLGRAPH (Portugal)

POLGRAPH was developed at the University of Aveiro, Portugal. It was designed for evaluating the impact of industrial pollutant releases and for air quality assessments. It is a Gaussian plume dispersion model for continuous, elevated point sources to be used on a local scale over flat or gently rolling terrain.

2.2.11 Other Gaussian Models

Other Gaussian dispersion models include CALINE3 (USA), (a Gaussian dispersion model designed to determine pollution concentrations at receptor locations downwind of highways), BLP (USA), CAL3QHC/CAL3QHCR (USA), the Complex Terrain Dispersion Model (CTDM) and the (US) Offshore and Coastal Dispersion Model (OCD).

2.3 Eulerian Models

Eulerian grid models simulate turbulent flows, chemical reactions and aerosol transport over an area by dividing the solution domain (airshed) in an array of grid cells. In the Urban scale the use of Eulerian grid models has been increasing over the past few years, as they provide a more realistic and comprehensive description of the urban atmosphere, compared to Gaussian models (Sokhi *et al*, 2006).

The driving imperatives for the development of the first Eulerian models was the need to understand and develop control strategies for the reduction of photochemical smog (ozone and nitrogen dioxide are the principle gaseous species). However, contemporary Eulerian Models are extended to enable modelling other species groups and a range of primary and secondary airborne pollutants. Because photochemical smog results from complex non-linear reactions of oxides of nitrogen and volatile organic compounds in the presence of strong sunlight, accurate simulation of the processes generally requires numerical solution of large systems of chemical equations (although less complex alternatives are also available).

Thus Eulerian grid models have the potential to provide concentrations fields for a large range of air pollutants, a moderate to high spatial and temporal resolution. Eulerian models are widely used in micro-scale simulations in the form of Computational Fluid Dynamics (CFD) codes, where the mass, momentum, energy, pollutant concentration and reactant transport equations are solved as a coupled system (see also section 2.5 below).

Widely used Eulerian Grid models include: ChemRange model (Scheringer, 1996, 1997), CHIMERE, the Community Mesoscale Air Quality (CMAQ) Model, REMSAD, CAMx (Environ, 2003), UAM-AERO, UAM-VPM, URM and CalGrid (Yamartino R. *et al*, 1989)

In the Lagrangian modelling approach, air concentrations are computed by summing the contribution of each pollutant puff that is advected through the grid cell as represented by its trajectory. In a Lagrangian model, modelling the growth of the pollutant puff or explicitly modelling the growth of a cluster of particles can simulate dispersion. The receptor-oriented and source-oriented approaches using Lagrangian particle models involve establishing trajectories backwards in time and to the source, or forwards from the emission point, respectively. Forward trajectories indicate where an air parcel will be transported, while backward trajectories describe the probabilities of its source. The impact at the receptor site may be expressed in terms of pollutant concentrations or deposition fluxes.

Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) (Draxler and Hess, 1998) has gained great popularity for studying pollution dispersion and source-receptor relationships, along with the models FLEXPART (Stohl *et al*; 1998, 2002b, 2003b; Stohl and Thompson, 1999), ATMOS (Arndt and Carmichael, 1995; Arndt *et al*, 1997, 1998), RAPTAD (Random Particle Transport And Diffusion) (Williams and Yamada, 1990), the Regional Atmospheric Modeling System Lagrangian Particle Dispersion Model (RAMS-LPDM) (Eastman et al. 1995), STILT (Lin *et al*, 2003), the Australian Lagrangian Atmospheric Dispersion Model (LADM) (Physick et al. 1992)and the British Nuclear Accident Model (NAME) (Maryon et al. 1991, UK MetOffice).

2.5 Computational Fluid Dynamics (CFD)

Computational Fluid Dynamics (CFD) is currently a very active area of research and development. CFD based dispersion models are regularly used to simulate the small-scale and complex flow patterns created in urban areas by large buildings and urban "street canyons".

CFD has a clear advantage over other types of modelling in that it is based on fundamental equations of physics, incorporates detailed three-dimensional geometry describing the terrain, and it includes localised environmental information in the form of boundary conditions. Therefore, models of transport and dispersion of airborne pollutants through urban street canyons are usually being developed using a computational fluid dynamics (CFD) approach.

The most fundamental consideration in CFD is how continuous fluids are treated in a numerical simulation. One method is by discretising the spatial domain into small cells to form a volume mesh or grid, and then applying a suitable algorithm to solve the equations of motion (Euler equations for inviscid, and Navier-Stokes equations for viscous flow.

The main difficulty is in relating turbulence mostly used in CFD with atmospheric scale turbulence drivern mostly by thermal convection rather than shear. The physical scale of CFD's application is mostly limited by the high computational cost. CFD models become more accurate as computing power becomes more widely available and inexpensive, and CFD models became capable of providing results in mesoscale dispersion situations and even in difficult cases, like complex terrain.

2.6 Empirical and Semi-empirical Deterministic Statistical Models

Empirical formulae are obtained statistically from experimental data sets. They provide a simple and convenient, but deceptive form of summarising extensive experimental data. As such, this technique is more suitable for relatively short-lived pollutants, such as NOx, or where the emission source density dominates the ambient concentrations.

Semi-empirical models incorporate deterministic elements, such as a Gaussian dispersion model, for specific components. Such an approach retains some of the elements of computational simplicity with the advantage of a deterministic approach to pollutant dispersion but sometimes has the disadvantage of uncertainties over the empirical components for future scenario or projections applications.

The main disadvantage of deterministic based models in evaluating and predicting the pollutant dispersion is their inability to predict the extreme concentrations (Khare and Sharma, 2002; Jakeman *et al.*, 1988). Also, lacking a rigorous connection with microstructure, these formulae do not offer predictive or interpretive power, seldom carry physical insight, and often fail when applied to a wider range of dispersion applications. Current statistical/empirical dispersion models are typically tuned to specific environments.

Moreover, the computational requirements of this technique are reasonably low.

2.7 Validation and Evaluation of Dispersion Models

Atmospheric dispersion models are an important tool to support decision-makers with spatial and temporal information on the dispersion forecast and the extent of the expected contamination. The accuracy of the calculations of spatial and temporal distributions of the air concentration, deposition and corresponding pollution levels is limited due to uncertainties in the applied dispersion algorithms, limitations in knowledge and availability of atmospheric input data, and the release conditions during an accident.

It is thus recommended that all models should be validated against well known experiments and any model evaluation exercise start with clear definitions of the evaluation goal and the variables to be considered. ETEX (Grazianni *et al.*, 1998) and the Model Validation Kit, are two widely recognised methods for evaluating long range dispersion models and are described in chapters 6.6.1 and 2.6.2. Other methods include ACURATE (Atlantic Coast Unique Regional Atmospheric Tracer Experiment), ANATEX (Across North America Tracer Experiment) and CAPTEX (Cross Appalachian Tracer Experiment).

2.7.1 The Model Validation Kit

The Model Validation Kit (Olesen, 2005) is a practical tool intended to serve as a common frame of reference for model performance evaluation and consists of four field data sets as well as software for model evaluation. The Kit has been used for the series of Harmonisation workshops and conferences (website: www.harmo.org).

A preliminary version of the Kit was used for the workshop in 1993, while a subsequent version was used essentially unchanged throughout the period 1994 - 2005 (in 1997, a supplement was added). The Kit was updated to Version 2.0 in October 2005. The new version allows the same studies to be carried out as the previous version, but has been revised in several respects. The package can be downloaded from the Internet at www.harmo.org/kit.

2.7.2 The European Tracer Experiment ETEX

The European Tracer Experiment (ETEX) (Dop, *et al.,* 1998) is a model evaluation method based on an experiment that was performed in October 1994 and involved two releases of a passive non-depositing tracer gas (perfluorocarbons) in western France and the subsequent dispersion over North Europe. The release was followed by tracer concentrations measurements for three days after the beginning of the emission using a sampling network spread over a large part of Europe. The sampling network consisted of 168 ground-level sampling stations in Western and Eastern Europe. To complement the meteorological measurements routinely gathered by the WMO network all over Europe, additional ground level and upper-air meteorological measurements at the release site were performed to obtain a comprehensive meteorological database (Dop, H.V. *et al.,* 1998). The experimental database contains ground and upper air meteorological observations and prognostic meteorological fields from the ECMWF (Gryning *et al.*, 1998; Straume and Nodop, 1997).

2.7.3 Validation of the proposed methodology

Both Lagrangian dispersion models used in this research have been extensively validated against experiments described above, hence no validation study was necessary in this study. FLEXPART has been evaluated using data from CAPTEX, ANATEX (Stohl et al., 1998) and the European Tracer Experiment (ETEX) (Stohl and Wotawa, 1997). HYSPLIT has as well been validated using CAPTEX (Draxler, 1987) ANATEX (Draxler, 1991) and ETEX (Graziani, 1998). Both models performed well in the above validation studies [e.g. (Stohl 1998); (Draxler, 1991)]. However further validation studies were performed in this study, comparing models' results with ground level measuremenst (e.g. Figure 5.25) and satellite data (e.g. Figure 5.27), which are described in chapter 5.

Chapter 3

3. The proposed modelling system

3.1 Overview

Particle dispersion models generally use meteorological fields and emission inventories as input, and calculate time-dependent concentration fields as output, using suitable models for the relevant processes such as advective or convective transport, mixing by diffusion, and mass transfer through dry or wet deposition, or chemical reaction (figure 3.1).

The applicability of Gaussian plume models is restricted by the assumptions on which the deduction of the mathematical equations for the Gaussian model from Newton's equations of motion is based. The most notable assumptions are constant emission rates, flat and homogeneous sites and a horizontally homogeneous wind field.

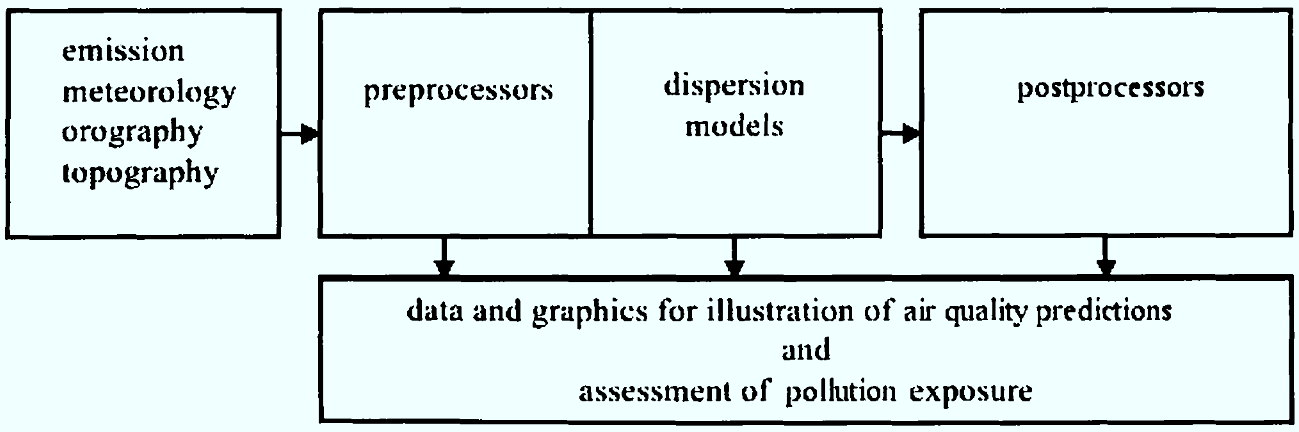

Figure 3.1 Typical atmospheric dispersion modelling system

Under these conditions, a Gaussian model may lead to unreliable results especially for short range applications, as long as the model is not adapted to these conditions; differences in the results of dispersion calculations using different modelling methods or flat / complex terrain have been shown in literature (Wichmann-Fiebig, 1999; Thehos *et al*, 1994).

1. Compared to the Eulerian methods, the Lagrangian method offers the following advantages:
2. No artificial numerical diffusion such as it occurs in Eulerian models. This is of special importance in the vicinity of the emission, where in Eulerian models the pollutant is instantaneously mixed over at least one grid box, which can cause large subsequent transport errors (A. Stohl *et al*, 2005);
3. Lagrangian models have the potential advantage to be simpler, more flexible, and computationally inexpensive;
4. Ability of modelling dispersion backward in time with Lagrangian particle models (Flesch *et al.*, 1995). Euler methods have difficulty in performing receptor based simulations in 3 dimensions. Adjoined solutions are needed but have only been tested in ground water dispersion;
5. Lagrangian models automatically achieve the mass conservation of the tracer gas, without extra computational needs;
6. The ability of Lagrangian models to easily handle emissions from point and line sources and the ability to calculate accurately the advection and dispersion from various emission sources;

7. Complete characterisation of the impact of turbulence on the transport of air pollutants and ability of Lagrangian models to incorporate more turbulence properties than the Eulerian models;

The disadvantages of Lagrangian models compared to Eulerian models are:

1. Eulerian models treat easily the chemical interactions of all air parcels within a grid square, or of pollutants that react non-linearly;
2. Eulerian models are more efficient than Lagrangian models in situations with a large number of sources;
3. A large number of particles is required in order to avoid sampling errors that occur when estimating pollutant concentrations by counting the number of particles in a volume.

This research uses and compares two widely used LPD models: the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model and the model FLEXPART. Both models possess the forward tracking and inverse (or receptor-based) modes. Apart from the above described advantages of LPD models, the Lagrangian method was selected for this research due to the availability of validated software such as FLEXPART and HYSPLIT and the free access to source code and an extensive user community of FLEXPART.

A detailed description of the Lagrangian particle models FLEXPART and HYSPLIT, can be found at (Stohl *et al*, 2005) and (Draxler, 1998) respectively. The models are also briefly described in chapters 3.1 (FLEXPART) and 3.2 (HYSPLIT).

LPD models generally require specific dispersion input parameters such as wind direction and wind velocity, roughness length. These data are usually obtained from separate pre-processor "weather" programs (figure 3.1), which have to be applied before the atmospheric dispersion can be simulated, or from meteorological services. The reference version of FLEXPART is driven with input meteorological data from the European Centre for Medium-Range Weather Forecasts (ECMWF) hence an ECMWF user account was required in order to operate the model.

This account enabled access to the MARS archive of ECMWF and retrieval of data from 1950 up to today, a 5-day forecast capability and variable resolutions that can reach a fine limit of 0.35 degrees latitude longitude and 90 vertical levers. In contrast, HYSPLIT has a pre-processor that enables input from various data sources, including ECMWF and the PSU/NCAR mesoscale model (known as MM5), hence the same data could be used for both models. To allow for more flexibility a weather model needed to be installed and operate locally on the workstations and the mesoscale PSU/NCAR model (MM5) was considered. Meteorological fields by meteorological datasets output from the MM5 mesoscale model or from datasets from the ECMWF MARS archive are used to initiate the MM5 model. Some of the advantages of MM5 over other weather data sources are:

- Ability of high-resolution simulations of weather phenomena (down to 1 km), since it is a mesoscale model.
- It is written in a non-hydrostatic framework of weather, meaning that it to produce more accurate forecasts for smaller localities
- Since the resolution is higher, forecasts for terrain, (when mountains are present), are more accurate

- It is widely used, hence user support is available through user forums and a big variety of input/output tools compared to other models (such as WRF).

While HYSPLIT allows the input of MM5 data, standard FLEXPART is only driven with fields from the ECMWF. Wotawa et al (2000) have developed and offered an early version (3.1) of the official FLEXPART (6.2 as for year 2006) that is driven by MM5 weather data. One of the duties of this research was to maintain the MM5 version of FLEXPART in order to import improvements included on later versions (6.2 in year 2006). This "maintained" version of MM5/FLEXPART became equivalent to A. Stohl's reference version 5.0 (2003) including updates such as the addition of a scheme for the parameterisation of moist convection (Seibert *et al.*, 2001; Emanuel *et al*, 1999) (version 4), better backward calculation capabilities (Seibert and Frank, 2004), and improvements in the input/output handling (version 5).

The outcome of this work carried out in this study is the modelling system used in the ATMMA project which comprises of:

- Two trajectory and dispersion modelling systems as are incorporated in the Lagrangian Particle (LPD) codes HYSPLIT (Draxler, 2001) and FLEXPART (Stohl *et al* 2005)

- A meso-scale weather model, namely MM5

- Input air quality data from the EXPER/PF, Air Quality Archive (website: http://www.airquality.co.uk), KentAir databases (website: http://www.kentair.org.uk) and Aerosol Index (satellite) data from TOMS (http://jwocky.gsfc.nasa.gov) and OMI (http:// http://aura.gsfc.nasa.gov/instruments/omi/index.html).

- Input meteorological boundary conditions provided by the ECMWF

- A web interface where partners can request a model run available at http://staffweb.cms.gre.ac.uk/~ps60/attma/models.htm

The main elements of the proposed modelling system are presented in Figure 3.2

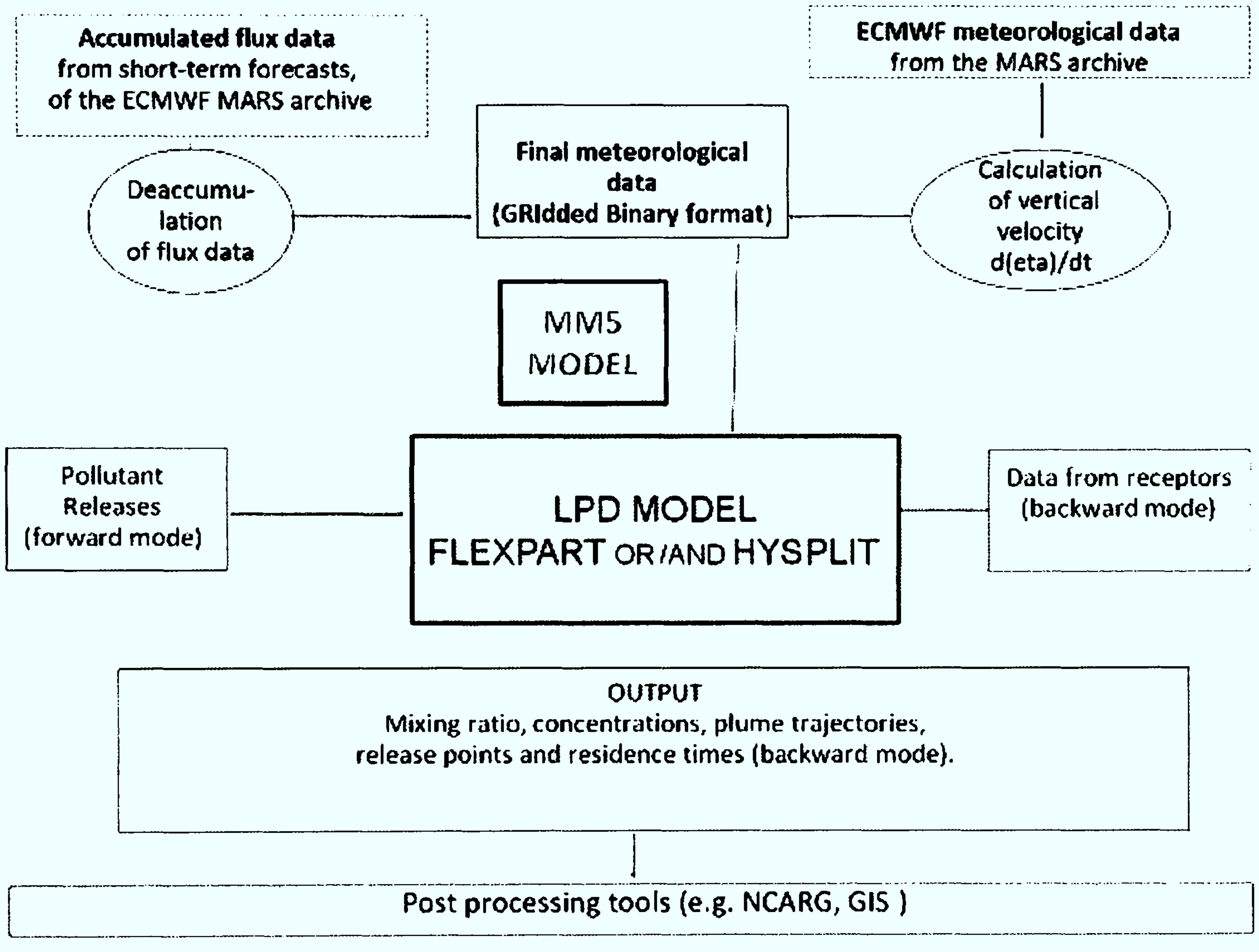

Figure 3.2 Schematic diagram describing the order of the main elements of the proposed modelling System and their primary functions.

3.2 Lagrangian Particle Dispersion models

3.2.1 Overview

This research utilises two widely used Lagrangian Particle Dispersion (LPD) models: the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model and the model FLEXPART. The advantages of FLEXPART and HYSPLIT over other LPD models are described in more detail in the following chapters.

HYSPLIT was developed by a joint effort between the National Oceanic and Atmospheric Administration (NOAA) and Australia's Bureau of Meteorology. The binary installation files are available for download from HYSPLIT's official website at NOAA (http://www.arl.noaa.gov/ready/hysplit4.html). Alternatively, HYSPLIT can be run interactively on-line at the same website.

FLEXPART is a model developed by Andreas Stohl (University of Munich). It is a community based open source LPD model, so available in source code format at the website: "FLEXTRA and FLEXPART homepage by Andreas Stohl and others" (http://zardoz.nilu.no/~andreas/flextra+flexpart.html).

Both models can operate in forward tracking and inverse (or receptor-based) modes.

3.2.2 LPD Model FLEXPART

LPD FLEXPART has been based on FLEXTRA, a kinematic trajectory model, also developed by Andreas Stohl (A. Stohl *et al*, 1995, 1998). FLEXPART is an open source project, freely available for scientific use and is widely used to calculate the transportation and dispersion of non-reactive pollutants in the atmosphere. The initial version of FLEXPART was designed to calculate the long-range and mesoscale dispersion of air pollutants from point sources. In later versions it evolved into a comprehensive tool for atmospheric transport modelling and analysis. FLEXPART is developed in FORTRAN77 programming language and comprises 144,517 lines of code (calculated with kloci tool, available at http://www.analogx.com/files/kloci.exe)

The main advantages of FLEXPART over other alternative LPD models are considered and described below:

1) FLEXPART's availability

FLEXPART is an open source project: Open source usually refers to software projects in which the source code is available to the general public for use and/or modification from its original design, free of charge. The code of FLEXPART is created as a collaborative effort in which programmers improve upon the code and share the changes within the community. Hence it can be modified, optimised and adjusted for each special case study. Since FLEXPART is being used and developed continuously by a growing user community (more than 31 user groups from 16 countries, in 2005). FLEXPART's community can share useful model code or code for the pre/post-processing of the input/output data. Generally open-source projects offer the following advantages:

- The important availability of the source code and the right to modify.

- The right to redistribute modifications and improvements to the code, and to reuse other open source code, permits all the improvements made to the code to be shared by large communities.
- Ability to perform a thorough inspection and verify the correctness of the algorithms and the implementation scheme used (not black box projects).
- There is no single entity on which the future of the software depends. The development can continue in the direction of the public demand, even if the original authors decide to discontinue or change the original aims of the project.

There are also serious disadvantages of open source code: Introduction of unfinished code, quality control of sections added by various users, non uniformity of coding style, errors introduced by users who do not really understand the original code, etc.

2) FLEXPART's code quality and maturity.

FLEXPART has been validated with measurement data from large-scale tracer experiments (Stohl *et al.*, 1998), during case-studies of stratospheric intrusions (Zanis *et al.*, 2003) and performs well in comparison with other similar models (Meloen *et al.*, 2003). It has been used for studying the intercontinental transport of ozone (Stohl and Trickl, 1999), the advection of Canadian forest fire emissions to Europe (Forster *et al.*, 2001), the dispersion of aircraft emissions in the stratosphere (Forster *et al.*, 2003), and has been used for inverse modelling in order to determine the source regions of North American pollution plumes measured over Europe (Stohl *et al.*, 2003), for example. FLEXPART has also been used to calculate a climatology of stratosphere-troposphere exchange (STE) processes, based on the **European Centre**

for Medium-Range Weather Forecasts (ECMWF) 15-year re-analysis dataset (ERA-15) (James *et al.*, 2003a,b). Furthermore, the community based nature of FLEXPART offers the advantage that as users have access to the source code, they can suggest improvements to the methodology and the code can be well documented and optimised for run-time performance. More information about validation kits (such as the ETEX, CAPTEX and the Model Validation Kit) is provided in chapter 2.7.

3) FLEXPART's portability

FLEXPART's source code can be compiled by a great number of FORTRAN compilers, including the open source g77 FORTRAN77 compiler of GCC. Hence it can be installed in most UNIX computers and potentially compiled on multiprocessor computers or computer clusters, or can be easily run remotely due to its simple command line interface. In this research FLEXPART was compiled on a PC Linux (x86) workstation, using the g77 compiler.

4) FLEXPART's modelling capabilities

Flexpart has a wide range of capabilities compared to other codes,
such as:

- Comprehensive modelling of physical processes, such as particle mixing within convective clouds.
- Ability of FLEXPART to operate with forecast data; FLEXPART also enables a "warm start" option, where a simulation can be reinitiated with updated meteorological data.
- Ability to calculate dry and wet deposition (Removal processes) of pollutants

- Clustered plume trajectories. Condensed particle output using a clustering algorithm (Stohl *et al*, 2001) can be outputted from FLEXPART.

- Ability to calculate uncertainties of the output from the standard deviation of the calculated concentration estimates.

The main disadvantages of FLEXPART, related to this research are:

1. **FLEXPART's dependency on meteorological fields from ECMWF.** The reference version of FLEXPART can only be driven by the proprietary meteorological fields provided by the European Centre for Medium-Range Weather Forecasts (ECMWF) and encoded under the Gridded Binary (GRIB) format. This makes the research dependent onto third party companies/organisations. Furthermore, the necessary linkage of FLEXPART to proprietary input libraries (ECMWF) required for reading Gridded Binary (GRIB) meteorological files, limits its usage to only the operating systems supported by the ECMWF (UNIX). A big effort had to be invested in this research to adapt FLEXPART for operation with alternative types of data, such as data provided by mesoscale models such as the MM5.

2. **FLEXPART's lack of features such as a Graphical User Interface (GUI) or a variety of pre-processing and post-processing facilities.**

The lack of choices in terms of post-processing utilities makes FLEXPART's adaptation a time consuming process, as new routines have to be developped for the analysis of the results. The lack of Graphical User Interface (GUI) makes FLEXPART difficult to be conFigured by users with low or medium expertise in computer technologies.

3.2.2.1 Input meteorological data fields

FLEXPART is an off-line model, driven by meteorological fields (analyses or forecasts) of the numerical weather prediction model of the European Centre for Medium-Range Weather Forecasts (ECMWF). The fields are defined by a hybrid, terrain following co-ordinate system encoded in Gridded Binary (GRIB) format. FLEXPART needs five 3-D fields: horizontal and vertical wind components, temperature and specific humidity, and the following two-dimensional fields: surface pressure, total cloud, cover, 10m horizontal wind components, 2m temperature and dew point temperature, large scale and convective precipitation, sensible heat flux, solar radiation, east/west and north/south surface stress, topography, land-sea-mask and subgrid standard deviation of topography.

Wotawa *et al* (2000) have developed MM5 driven versions of the trajectory model FLEXTRA and the particle diffusion model FLEXPART based on the reference version of FLEXPART version 3.1 (reference version for the year 2000 when the MM5 version was developed). The MM5 version makes FLEXPART capable of analysing transport on scales from global down to local scale such as plumes from cities and large power plants.

For the needs of this research, Wotawa's MM5 FLEXPART was updated (with the kind support of Brett Anderson, US EPA) to improvements made on the code of FLEXPART by Stohl *et al* after the year 2000 (FLEXPART 3.1). Our updated version of MM5 FLEXPART became equivalent to A. Stohl's original version 5.0 (2003) including updates such as the addition of a convection scheme (Seibert *et al.*, 2001), (Emanuel *et al*, 1999) (version 4), better backward calculation capabilities (Seibert and Frank, 2004), and improvements in the input/output handling (version 5).

Caroline Forster (Norswegian Institute for Air Research, Norway) in 2005 ported FLEXPART's Version 6 to input weather data from the GFS model (Kalnay and Kanamitsu, 1988). Also, due to its open-source nature, FLEXPART was the first model able to operate with data from the WRF Mesoscale Model (Janjic, *et al* 2004), ported by Jerome Fast (Pacific Northwest National Laboratory, USA). The interface for the ALADIN Numerical Weather Prediction Project (ALADIN International Team, 1997) has also been developed by Helfried Scheifinger and Mathias Langer (Zentralanstalt fuer Meteorologie und Geodynamik, Austria)

3.2.2.2 Vertical motion

Data from the ECMWF model are defined by a hybrid terrain following coordinate system and give no direct information on the vertical component of the wind velocities. The vertical wind in hybrid coordinates is calculated mass-consistently from horizontal wind data by the pre-processor (see data preparation chapter).

3.2.2.3 Atmospheric Boundary Layer (ABL) parameters

The Atmospheric Boundary Layer (ABL) heights are calculated according to Vogelezang and Holtslag (1996), based on the concept of critical Richardson number. The ABL height h_{mix} is set to the height of the first model level *l* for which the Richardson number R_{il} exceeds the critical value of 0.25 :

$$R_{il} = \frac{(g/\Theta_{v1})(\Theta_{vl} - \Theta_{v1})(z_l - z_1)}{(u_l - u_1)^2 + (v_l - v_1)^2 + 100 u_*^2} \qquad (5)$$

Where Θ_{v1} and Θ_{vl} are the virtual potential temperatures, z1 and z*l* the heights of, and (u1, v1), and (u*l* , v*l*) are the wind components at the heights and u_* is the friction velocity.

3.2.2.4 Moist convection

FLEXPART implements a parameterisation of convective mixing. As the model is driven by meteorological fields from the **European Centre for Medium-Range Weather Forecasting** (ECMWF), which has no direct information on convection, a convection scheme is implemented to derive the convective redistribution of air from the resolved scale fields of temperature and humidity. This was initially implemented in FLEXPART by Siebert, *et al.* (2001) with the scheme of Emanuel and Zivkovi´c-Rothman (Emanuel et al. 1999), a physically-based development that describes the effects of updrafts and downdrafts with entrainment, detrainment and compensating subsidence. The scheme produces an Eulerian redistribution matrix which is used for a stochastic redistribution of particles. When activated in the simulations, the moist convection scheme, usually accounts for 70% of the overall computational needs of FLEXPART (Stohl *et al*, 2005).

3.2.3 HYbrid Single-Particle Lagrangian Integrated Trajectory model (Version 4)

3.2.3.1 Overview

The HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT_4) model (Version 4) was used in this research as an alternative formulation to the LPD model FLEXPART. HYSPLIT is a result of the collaboration between the US National Oceanic and Atmospheric Administration (NOAA) and Australia's Bureau of Meteorology and, like FLEXPART, is able to compute particle dispersion and deposition simulations as well as trajectories. HYSPLIT is a closed source code that contains a complete Graphical Unit Interface developed in Tcl/Tk and output graphical facilities. In this respect it is a more complete product for the user than FLEXPART. It can be used either via the on-line interface of NOAA Air Resources Laboratory's (ARL) website or via the downloadable installation (binary) software. It is available at no charge to individual users who have a formal affiliation with one of the institutions engaged in atmospheric sciences or in the provision of atmospheric operational products, and for governmental, educational (as in the case of this research), or non-profit use. Compared to FLEXPART, HYSPLIT offers more options for the dispersion method that can be based either on puff or particle approaches. HYSPLIT is optimised to input meteorological model forecast data from the National Center for Environmental Prediction (NCEP) archived by the Air Resources Laboratory (ARL) on one of three conformal map projections (Polar, Lambert, Mercator). However, it provides several tools to convert data from various sources including MM5 and ECMWF into the ARL format. The dispersion rate in HYSPLIT is calculated from the vertical diffusivity profile, wind shear, and horizontal deformation of the wind field (Draxler *et al*, 1997). Air concentrations are calculated at a specific grid point for puffs and as per cell-averaged concentrations for particles.

An important feature of HYSPLIT, which has been used in this research, is the desert dust emission module. The emission module of HYSPLIT is based on the concept that threshold friction velocity is dependent on surface roughness. Surface roughness then is correlated with soil properties. A dust emission rate is computed from each cell of an emission matrix, when the local wind shear exceeds the threshold velocity for the soil characteristics of that emission cell. The emitted material is dispersed and transported using HYSPLIT.

A) The following advantages of HYSPLIT were considered prior to its introduction in this research:

- **Code maturity and quality**

  HYSPLIT is one of the most popular Lagrangian models and it has undergone many improvements, since its original version

- **Ease of use**

  Unlike FLEXPART, HYSPLIT is provided by NOOA as a complete package, easy to install (binary installation files for Windows and Macintosh) and accompanied with a Graphical Unit interface and various tools to input data and analyse the results

- **Computational Efficiency.**

Benchmark comparisons between FLEXPART and HYSPLIT in terms of computational efficiency are limited by the closed source availability of HYSPLIT, so that both models cannot be compiled by the same compiler and perform under the same operating systems. However our general experience is that HYSPLIT performs very efficiently, compared to FLEXPART. It is able to model the same case using less computational resources, compared to FLEXPART which can be attributed to the convection scheme that generally counts for the 70% of the overall CPU usage in FLEXPART (Stohl, 2005).

- **Availability**

HYSPLIT is free-ware for personal use in environmental research.

- **Many choices for handling dispersion**

3-D particle dispersion or splitting puffs (top-hat or Gaussian)

- **Availability of several tools to input data and analyse the results.**

HYSPLIT is provided with a great variety of tools to input meteorological data from various sources (including ECMWF) and post-processing utilities (converter to GIS, plotting facilities etc)

- **Special simulation modules such as the desert dust emission module, recently revised for more accurate predictions.**

Transcontinental dust transport episodes from Africa are a cause of many exceedences of particulate matter in Western Europe. In this study we used this feature to model dust storms affecting the air quality of Europe, originating in the Sahara desert.

B) Disadvantages of HYSPLIT, considered in this research include:

- **Closed source format**

Although HYSPLIT's code can be available to registered users, the model is under a licence scheme that does not allow for modifications in the source code.

- **Lack of a sophisticated methodology for moist convection and vertical motion parameterisation**

HYSPLIT does not take into account the vertical (convective) mixing, an important transport mechanism compared to other Lagrangian models such as FLEXPART. As we will see in the results, this leads to significant differences between the two codes, when vertical distributions are considered.

3.2.3.2 Input of Meteorological Data Fields

HYSPLIT uses meteorological model forecast data from the NCEP archived data in the ARL format. However in order to enable the input of different data sources, meteorological profiles are always pre-processed and at each horizontal grid point are linearly interpolated to an internal dispersion model terrain-following (□) coordinate system. The dispersion model's horizontal grid system is then identical to that of the meteorological data and three different conformal map projections are supported: Polar Stereographic, Mercator, and Lambert Conformal, using a set of universal mapping transformation routines (Taylor, 1997).

This enables HYSPLIT to provide several tools for converting data from various data sources, including fields from the ECMWF. Meteorological data fields that may be provided on one of the following four different vertical co-ordinate systems: pressure-sigma, pressure-absolute, terrain-sigma, or a hybrid absolute-pressure-sigma (used by

ECMWF). HYSPLIT needs U, V (the horizontal wind components), T (temperature), Z (height) or P (pressure), and the pressure at the surface, P_o. Moisture and vertical motion are optional; however the vertical motion may be computed based upon how the vertical co-ordinate is defined (see 3.3.3.2). The time interval between the meteorological fields should be constant for the gridded fields. If wet deposition processes are to be calculated, HYSPLIT also requires the rainfall field.

HYSPLIT can directly use meteorological fields from the following sources:

Forecast Data

ARL server for HYSPLIT compatible (ARL format) data

ARL ASEAN is a low-res ARL formatted file extracted for Asia

NCEP GFS global GRIB converted to ARL format

NCEP NAM mesoscale GRIB converted to ARL format

Analysis Data

ARL current format archives for the last two days

ARL archive of long-term ARL formatted data

NOAA reanalysis data archive from the ARL server

NCEP GDAS converted to ARL format

Alternatively HYSPLIT can import data from the following sources (and then convert to the ARL format):

NAM forecast mesoscale GRIB files

NDAS archive analysis GRIB files

GFS forecast global GRIB files

GDAS archive global GRIB files

ECMWF forecast global GRIB files (≤ 1.0 deg)

ECMWF archive global ERA-40 GRIB files

MM5V3 regional Version 3 binary output files

MCIP IOAPI formatted files

User entered single station and level user data entry

3.2.3.3 Vertical Motion

When the input meteorological data contains a vertical motion field (usually in pressure units), the vertical velocity field is almost always relative to the meteorological model's native terrain-following sigma co-ordinate system and can be used by the trajectory and dispersion model calculations, after it is remapped to a common vertical co-ordinate system.

However when meteorological data do not directly provide fields of the vertical motion, such as data from the ECMWF, the dispersion model replaces these fields with an internally calculated vertical velocity based upon an assumption that the pollutant parcel is transported on some other surface. The input data can be remapped to various surfaces by computing the velocity (z) required to maintain a parcel on the selected (η) surface,

$$W\eta = \frac{(-\frac{\partial \eta}{\partial t} - u\frac{\partial \eta}{\partial x} - u\frac{\partial \eta}{\partial y}}{\frac{\partial \eta}{\partial z}} \qquad (13),$$

given the slope of the surface and its local rate of change and where the surfaces, η, can be either isobaric (p), isosigma (σ), isopycnic (ρ), or isentropic (θ) (Draxler 1997).

The way that HYSPLIT handles vertical motion is a key difference to the methodology of FLEXPART and is assumed to be a major reason for the differences observed between the output results of the two models. FLEXPART calculates in its pre-processor the vertical wind component in hybrid co-ordinates, mass-consistently (i.e. to satisfy mass continuity) from spectral data (see 3.2.1.3).

3.2.3.4 Atmospheric Boundary Layer (ABL) Parameterisation

HYSPLIT assumes the ABL depth at each grid point to be equal to the height at which the potential temperature first exceeds the value at the ground by 2K. To determine the boundary layer depth, the temperature profile is analysed following a top to bottom approach, to reduce the influence of possible shallow stable layers near the ground. Night-time PBL depths are expected to be overestimated (Draxler, *et al*, 1997). The height is chosen to correspond with the minimum height resolution typical of the meteorological input data.

3.2.3.5 Dry and Wet Deposition handling in HYSPLIT

HYSPLIT can calculate dry deposition using details about the nature of the surface, or a constant velocity can be defined explicitly. Gravitational settling of particulate matter can also be calculated from Stokes law, using user data for particle shape, size and density. In addition to the mass removal option,

Wet deposition occurs when pollutants are deposited in combination with precipitation, predominantly by rain and snow, but also by clouds and fog. T HYSPLIT parameterises both the within-cloud (washout) and below-cloud (rainout) scavenging. If the winds are sufficiently strong, and the pollutant is not bound to the surface, then re-suspension can also occur.

In the case of nuclear incidents, radioactive decay is incorporated (Draxler, *et al*, 1997).

3.2.3.6 HYSPLIT's desert dust emission algorithm

The emission module of HYSPLIT is based on the concept that threshold friction velocity is dependent on surface roughness. Surface roughness then is correlated with soil properties. The revised dust emission module (Westphal *et al*, 1987) was incorporated into HYSPLIT starting with version 4.6. In the revised version of the emission module, the flux (g/m²) equation:

$$F = 0.01 u_*^4 \tag{14},$$

used by Westphal et al. (1987) replaced the Marticorena equation where vertical mass flux (F) is calculated from the friction velocity and the threshold friction velocity:

$$F = K \,\square\, g^{-1} u_*(u_*^2 - u_{*_t}^2) \tag{15},$$

Where K is the the soil texture coefficient found by Gillette et al. (1997) and has a value of $5.6 \times 10^{-4}\ m^{-1}$ for sand soils. In both approaches, dust emissions only occur during dry days when the friction velocity exceeds the threshold value (0.28 m/s for an active sand sheet). Over the typical range of wind speeds that result in dust emissions, the emission flux is about a factor of 10 lower using the Westphal equation (Draxler 2004).

The emission domain specified by the user, is divided into one-degree latitude-longitude grid cells and the points found to have a desert land-use category are set as possible emission points (409 in the case of Sahara). Dust particles are only emitted from those cells when the wind speed exceeds the emission threshold associated to the soil characteristics of the emission cell (Westphal *et al*, 1987). Finally, the emitted material is dispersed and transported using the standard Lagrangian methodology of HYSPLIT (Draxler and Hess, 1998).

The methodology can be applied to predict the arrival of a dust cloud, if forecast data are used.

3.2.3.7 Validation

HYSPLIT has been validated against air concentration results from the European Tracer Experiment (ETEX) and also compared to 27 other operational ATM models. There were two phases to the second evaluation. (NOAA, 1997: Draxler *et al*, 1997). In both these evaluations, HYSPLIT results generally were in the middle of the performance range, in the realtime phase due to coarser resolution of Aviation (AVN) Global Model forecasts over Europe compared with the data fields available to the European NWP centres and in the post-experiment phase due to pre-processing the ECMWF data at a resolution to be consistent with the previous real-time simulation (NOAA, 1997: Draxler *et al* 2002). More information regarding validation methods of atmospheric dispersion models can be found at chapter 2.7 .

3.2.4 General Comparison of LPD models FLEXPART and HYSPLIT

The following table summarises the main differences between the two LPD models:

Table 3.2 Comparison of LPD models FLEXPART, HYSPLIT and UK NAME

Model Name	**HYSPLIT**	**FLEXPART**	**UK Nuclear Accident Response Model (NAME)**
Latest Version (as of May 2006)	4.7	6.2	NAME III
Developer	R. Draxler (NOOA, USA)	A. Stohl (NILU, Norway), prev. University of Munich	UK's Met Office
Availability	Freeware from NOOA website (http://www.arl.noaa.gov/ready/hysp_info.html)	Open Source (Freeware), from A. Stohl's Flextra/Flexpart website: http://zardoz.nilu.no/~andreas/flextra+flexpart.html	Property of the UK Met Office
Source code availability	Only for registered users and offered without modification permission.	YES	No
Programming Language	FORTRAN 90	FORTRAN 77	FORTRAN 90
Basic Software Requirements	Internet connection and www browser for the online version, or Windows 95/2000/XP and Apple Mac OS.	Any (UNIX) computer with Fortran 77 compiler and GRIB decoding libraries.	UNIX workstation
User Interface	Console Mode or Tcl/Tk Graphical Unit Interface (GUI) with	Console Mode	Console mode

Model Name	HYSPLIT	FLEXPART	UK Nuclear Accident Response Model (NAME)
	integrated html compatible help.		
Dispersion method	3-D particle, puff, or hybrid.	3-D particle.	Puff when modelling dispersion over a short range
Initial Diffusion	Gaussian, top-hat or from a fixed particle position.	Gaussian or from a fixed particle position.	Using random walk techniques
Meteorology	ARL data from NCAR/NCEP (natively) or converted from NAM, GFS, ECMWF, MM5, RAMS, COAMPS, and other data	GRIB data from ECMWF (also MM5 and AVN only for FLEXPART version 3.2)	3-dimensional meteorological data from the Met Office's Unified National Weather Prediction Model.
Special Simulation	Chemistry Conversion Module, Module for Emissions from Dust Storms	Temporal variations of emissions (Since version 6.0)	Rise of buoyant plumes, plume chemistry focusing on sulphate and nitrate chemistry, European Radar Rainfall (ERR):
Vertical motion when not available from met data	Not calculated, methodology based upon the assumption that pollutant parcels are transported on some "other" surface.	Calculated by the pre-processor, based on mass continuity.	Available from Unified model data

Model Name	**HYSPLIT**	**FLEXPART**	**UK Nuclear Accident Response Model (NAME)**
Output Format	Concentrations in ASCII or FORTRAN unformatted files	Tracer concentrations and/or mixing ratios (for forward runs), or emission sensitivity response functions (for backward runs) Binary format	Tracer Concentration
Output trajectories	Available	Plume trajectories, using cluster analysis (Stohl *et al.*, 2002)	Available
Post Process/Analysis tools provided	Converters to ASCII text, postscript (using psplot), GIF, GrADS, GIS, ArcView (GIS), Vis5D and NCARG . Also various source attribution tools such as Source--Receptor Matrices.	Converter to NCAR Graphics	N/A

Table 3.2 Comparison of LPD models FLEXPART and HYSPLIT(cont.)

3.3 Input Meteorological Fields

Air pollution simulations with LPD models generally use meteorological fields and emission inventories as input, and calculate time-dependent concentration fields as output, using suitable models for the relevant processes, such as transport, diffusion, deposition. The models in this research were driven by meteorological datasets from the European Centre for Medium-Range Weather Forecasts (ECMWF) and the MM5 mesoscale model, hence they are briefly described in the following sections.

3.3.1 The European Centre for Medium-Range Weather Forecasts (ECMWF)

3.3.1.1 Overview

The European Centre for Medium-Range Weather Forecasts (ECMWF) is the result of a joint effort between several European meteorological services and the Centre has been established with the goal of making medium range (5-10 days) forecasts with a global coverage. Two types of data obtained from the ECMWF have been used in this research: Operational and Re-analysis data, provided by the ERA-15 and ERA-40 projects. The new re-analysis project ERA-40 covers the period from mid-1957 to mid-2002 overlapping the earlier ECMWF re-analysis, ERA-15, 1979 to 1993. The data sets are based on quantities analysed or computed within the ERA-40 data assimilation scheme or from forecasts based on these analyses. The data sets are so-called analyses, which is a combination of observations and numerical calculations. Measurements from satellites, radio sondes, weather stations, etc. are assimilated into a meteorological model, that produces an estimate of the state of the atmosphere at a given time. An analysis is used as starting point for the production of weather forecasts.

Although ECMWF web services currently give to non-members immediate and free access to ECMWF 15 and 40 Years Re-Analysis datasets (at a resolution of 2.5 x 2.5 degrees), an authorised user account is essential for access to the ECMWF computing facilities (ECGATE , Mars archive). Such access enables the preparation and retrieval of high resolution data (up to higher than 0.5° longitude and latitude, 60 vertical levels and frequency of 3 hours) that can be used as a direct input for FLEXPART. The data are available for period 1950 until today and 5 days forecast. Since December 2003, this research project has been granted a full user account and full access is enabled to ECMWF's computing facilities, such as the Mars archive.

3.3.1.2 The ECMWF GRIded Binary data packing format

ECMWF uses a modified version of the GRIB format for storing meteorological data. GRIB (WMO, 1998) (designated FM 92-VIII Ext. GRIdded Binary) is the general purpose data exchange format, approved by the World Meteorological Organization (WMO) Commission for Basic Systems (CBS) Extraordinary Meeting Number VIII (1985). Although it was designed as a standard for data storage many organizations use their own versions, as for example ECMWF.

By packing information into the GRIB code, records (data) can be made more compact than character oriented bulletins, which will produce faster computer-to-computer transmissions (WMO, 1998).

The GRIB code form represents numeric data as a series of binary digits. Such data representation is independent of any particular machine representation; by convention data lengths are measured in octets. Data are coded as binary integers using the minimum number of bits required for the desired precision. Numeric values may first be scaled by a power of ten to achieve an appropriate decimal precision, a reference value is subtracted from them to reduce redundancy and eliminate negative values, and they may then be further scaled by a power of two to pack them into a pre-selected word length. The two scaling operations are independent; which, or both, are used in any given case depending upon choices made as to the method of packing. See below.

The representation of a single value is such that:

$$Y \times 10^{D} = R + (X \times 2^{E}) \qquad (16)$$

where:

Y = original or unpacked value;

D = decimal scale factor, to achieve desired precision (sign bit, followed by a 15-bit integer);

R = reference value (32 bits);

X = internal value (number of bits varies for each record);

E = binary scale factor for variable bit word length packing (sign bit, followed by a 15-bit integer).

The reference value (R) is the minimum value of the (possibly) decimally scaled data that is being encoded.

If second order (or "complex") packing is used the internal value, X, will be made up of two values, a "local minimum value", Xi, and a "second order packed value", Xj. There will be one Xj for each grid point and a variable number of Xi values.

The ECMWF GRIB data format description, along with the ECMWF-specific parameter table, and a list of differences between the WMO and the ECMWF versions of GRIB can be found at:

ftp://ncardata.ucar.edu/datasets/ds111.2/format

3.3.2 The Fifth-Generation NCAR / Penn State Mesoscale Model (MM5)

The model, known as MM5 is an open-source project and is continuously being improved by contributions from users at several universities and government laboratories. It is supported by several auxiliary programs, which are referred to collectively as the MM5 modelling system and can be compiled for most UNIX machines.

The PSU/NCAR Mesoscale Model is a community, limited-area, non-hydrostatic or hydrostatic (Version 2 only), terrain-following sigma-coordinate model designed to simulate or predict mesoscale and regional-scale atmospheric circulation. It can provide a good base for our simulations as it can output meteorological data of higher spatial and temporal resolution than the ECMWF fields. MM5 is the fifth generation of the model, originally developed by Anthes at Penn State in the early 70's (Anthes, *et al* 1978), and at NCAR (Grell, *et al* 1994).

Since the original version at Penn State, it has undergone many changes, including:

1. Multiple-nested grid mode,
2. Non-hydrostatic dynamics, which allows the model to be used at a few-kilometre scale,
3. Multi-tasking capability on shared- and distributed-memory machines,
4. A four-dimensional data-assimilation capability, and
5. More physics options, such as precipitation, planetary boundary layer (PBL) process parameterization and atmospheric radiation schemes.

1) Brief Description of the MM5 model

As in most mesoscale weather models, atmospheric dynamics within MM5 are simulated by computations carried out on one, or a set of horizontal grids, which span a specified geographic region (often called domain) and have a given grid resolution. The model performs calculations at each grid point based on physical relationships which govern atmospheric behaviour. Hence, the future properties of a particular point are provided by the combination of its current properties and those of the surrounding points, calculated using equations which relate these properties. Through repetition of this process, the model moves the state of the atmosphere forward in time, producing a weather forecast.

MM5 uses a terrain-following vertical co-ordinate system and has the capability to run with several horizontally nested domains (grids). This allows the model to run at a relatively high resolution over a specific area of interest, while maintaining a low resolution over the surrounding areas. This flexibility means that high-resolution forecasts can be produced for a smaller region in a much shorter time than would be possible if the entire large domain had to run at the fine grid spacing (because more points mean more computation time), while still allowing the model to simulate the larger-scale circulation.

isobaric meteorological data are horizontally interpolated from a latitude-longitude mesh to a variable high-resolution domain on either a Mercator, Lambert conformal, or Polar Stereographic projection.

The programmes RAWINS and little_r can enhance the interpolated data with observations from the standard network of surface and rawinsonde stations.

The vertical interpolation from pressure levels to the sigma co-ordinate system of MM5 is performed by the program INTERPF. Sigma surfaces near the ground closely follow the terrain, and the higher-level sigma surfaces tend to approximate isobaric surfaces. Since the vertical and horizontal resolution and domain size are variable, the modelling package programs employ parameterised dimensions requiring a variable amount of core memory. Some peripheral storage devices are also used.

3.4 Input Air Quality Data

FLEXPART and HYSPLIT make use of the chemical and physical characteristics of gases and particles measured at source and/or receptor to both identify the presence of and to quantify source contributions to receptor concentrations. The main sources for receptor air quality data in this research are the EXPER/PF database and the UK National Air Quality Information archive.

3.4.1 The UK National Air Quality Information archive

The UK National Air Quality Information archive was developed by NETCEN, part of AEA Technology Environment, on behalf of the UK Department for Environment, Food & Rural Affairs and the Devolved Administrations. The online interface to the archive (available at the website: http://www.airquality.co.uk) provides up-to-date, comprehensive, detailed information on current air quality. However, the site is also the national archive of air quality information and reports, including detailed air quality monitoring data and statistics and sections on local air quality management and air quality research.

3.4.2 The database of the EU-Interreg III "EXPER/PF" project

3.4.2.1 Overview

This research project, since February 2004, has been granted access to the atmospheric pollution database of "EXPER/PF" (Exposure of communities living in the centre of the Euro region to polluting atmospheric particles: the case of fine particulate matter). EXPER/PF European project (« Exposition des populations de l'Euro Région aux polluants atmosphériques: le cas des particules fines » - Population Exposure in Euroregion to Atmospheric Pollutants: case of Fine Particles) was launched in January 2002, within the framework of Interreg III program. Project's Objective is to develop and promote a cross-border (North of France-Flanders) database on atmospheric Particulate Matter, usable by the professionals involved in Public Health, Environment and Regional Planning.

3.4.2.2 Database contents

The EXPER/PF data base contains three types of data (Schadkowski, 2004):

1) routine in-situ pollutants measurements, provided by the fixed stations of the air quality networks which are project partners:

   Three air quality networks take part in the development of this data base: AREMA LM, OPAL'AIR and VMM. The measurements data are supplied on a one hour basis. The base was fed by the following data:

   - time measurements of PM10 et PM2.5
   - time measurements of gas compounds (CO, SO2, NOx, O3…), complementary of PM measurements.

3.4.3 Other Atmospheric Data sources

Other atmospheric and air quality data sources include:

1. The Kent and Medway Air Quality Monitoring Network

The Kent and Medway Air Quality Monitoring Network (accessed at: www.KentAir.org.uk) is the website of the Kent and Medway Air Quality Monitoring Network. The network is funded by the districts and boroughs within the county with an additional contribution from Kent County Council. The aims of the network are to promote the improvement of air quality within the region, help local authorities to meet their obligations under Environmental Regulations and maintain an accessible database of robust measurements for public reporting, research and development.

2. The British Atmospheric Data Centre (BADC)

The British Atmospheric Data Centre (BADC) is the Natural Environment Research Council's (NERC) designated data centre for the atmospheric sciences. The role of the BADC is to assist UK atmospheric researchers to locate, access and interpret atmospheric data and to ensure the long-term integrity of atmospheric data produced by NERC projects.

3. SEIPH - Environmental Research Group

Air Quality monitoring networks run by the SEIPH on behalf of London, Bedfordshire, Kent and Hertfordshire in the UK are reported. Near real-time data is available from the project's web site (http://www.londonair.org.uk)

4. Aerosol Index (Satellite) data from NASA's TOMS and OMI instruments

Aerosol index data from TOMS and OMI were used for validation of the output results.

3.5 Preparation and Installation of FLEXPART

In order to access ECMWF GRIB data, FLEXPART has to be linked to external GRIB encoding/decoding libraries. Also to obtain GRIB data from the ECMWF Mars archive, applicable as a direct input to FLEXPART, the (authorised) Mars users need to install the data extraction routines prepared by Gerhard Wotawa and Paul James. The following software was chosen for the procedures of modelling and data visualisation:

- Linux Operating system (SuSE 9.0) with the Fortran compiler GNU g77 (version 3.3.1) running on an x86 series processor (FLEXPART may also run on SUN, SGI, HP, Compaq Alpha workstations).
- FLEXPART version 5.0 source code, available at the official FLEXPART website maintained by Andreas Stohl:

http://www.forst.uni-muenchen.de/EXT/LST/METEO/stohl/FLEXPART.html

- The ECMWF GRIBEX software for decoding ECMWF re-analysis meteorological datasets encoded under the WMO FM-92 GRIdded Binary (GRIB) format.
- Ecdocs routines for extraction of ECMWF meteorological data, developed by Gerhard Wotawa *et al* (2000) available at: http://www.forst.uni-muenchen.de/EXT/LST/METEO/stohl/FLEXTRA/ecmwf_extr.html)
- NCAR Graphic libraries to visualise/plot the model's output available at: http://ngwww.ucar.edu
- Vis5D+ from http://vis5d.sourceforge.net
- Post-processing tools for the LPD Model's output.

GNU/Linux Operating System (distribution: SuSE 9.0) was the choice for this project, as a well established and free UNIX operating system. Other reasons for the choice are the ability of the LINUX OS to operate on a wide variety of hardware (e.g. laptops with x86 processor), its compatibility with the majority of the required tools (e.g. FLEXPART, NCARG, ECMWF data tools) and the availability of several open-source tools (e.g. G77 FORTRAN compiler, image converters etc).

The computer workstation that hosted the Particle Dispersion Model simulations and data visualisation is an x86 series Personal Computer powered by an Intel Pentium M 1.7 GHz processor and 1GB of RAM.

3.5.1 Preparation of ECMWF GRIdded Binary (GRIB) data, prior to July 2004

This paragraph provides a detailed technical description and a step-by-step setup guide for original routines developed Gerhard Wotawa and Paul James for the preparation and retrieval of ECMWF GRIdded Binary data. The routines are freely available at Paul James' web site: http://www.forst.tu-muenchen.de/EXT/LST/METEO/stohl/FLEXTRA/ecmwf_extr.html

These routines are no longer compatible with the ECgate system of ECMWF (after July 2004) and are no longer supported or maintained by G. Wotawa. The new version of retrieval routines by G. Wotawa, *et al* in July 2004 (see section 3.4.2) were developed entirely from scratch, hence they have different methodology and require a different setup procedure.

However, this paragraph can be particularly useful for FLEXPART users who want to pre-process data using our own data retrieval routines available at the website of our research project (http://staffweb.cms.gre.ac.uk/~ps60/attma) which are mostly based on the original routines of Wotawa but have been ported to the new ECgate server. As can be seen in paragraph 3.4.3, these maintained versions of the original routines offer more flexibility over the revised version of G. Wotawa routines. Also, this paragraph describes the data preparation procedure followed in many of the case studies presented.

The routines perform the following tasks:

- Retrieval of the accumulated flux data from the MARS archive (e.g. precipitation, radiation, sensible heat flux),

- De-accumulation of flux data, storage of results in the ecfs file-system,
- Retrieval of other meteorological fields
- Calculation of the vertical velocity component d(eta)/dt

The following paragraphs give a step-by-step guide to the above procedure.

After the routines are uploaded to the home user directory of the remote file system of ECMWF, they have to be extracted and conFigured to reach the correct paths to the account's directories. Also the type of required data should be conFigured, such as the region's co-ordinates and the beginning/ending date of the simulation. The routines for tasks 1 and 2 can be found in any of the directories:

fluxjobs_4dvar, fluxjobs_era (for) , fluxjobs_gen.

In each case the user has to choose which of the above directories corresponds to the required data type and period, for example the routines at fluxjobs_era prepare data of the ERA-15 type. For data from year 2000 and beyond, the directory fluxjobs_4dvar can be used and hence it was the preferred one for the project's simulations.

1) Retrieval of the accumulated flux data

First new directories should be created in the scratch filesystem to accommodate the flux data. This can be done after the user has connected to the remote ECgate system by the UNIX command:

mkdir $SCRATCH/flux

The FORTRAN include file "parameter" has to be modified during each data retrieval, according to the specifications of the required grid, such as:

nx, ny: number of grid points

xlon0, ylat0: longitude and latitude of lower left grid point

dx, dy: grid distance

Also the following scripts have to be conFigured: surfjob.new for the retrieval of the accumulated flux data from the MARS archive and deacc.flux for the deaccumulation of the fluxes. Then all the routines have to be compiled remotely with the FORTRAN compiler using the command:

f77 *.f $EMOSLIB

Then the binary output has to be renamed to FLUXACC , so that it can be run later by the scripts:

mv a.out FLUXACC

After all configurations are completed, the script surfjob.new is put in batch queue by executing the command:

qsub -q ecgate1.normal surfjob.new

The following flux data, required by FLEXPART, can be retrieved on a user-selectable geographical grid (lamda-phi) (Wotawa, *et al* 2003):

Abreviation	Type of Data	Unit
LSP	Large Scale Precipitation	mm/hr
CP	Convective Precipitation	mm/hr
SSHF	Surface sensible heat flux	W/m^2
EWSS	East-west surface stress	N/ m^2
NSSS	North-south surface stress	N/ m^2
SSR	Surface solar radiation	W/ m^2

Table 3.3 Flux data required by FLEXPART

2) De-accumulation of FLUX data

When the accumulated flux data is retrieved from the MARS archive, the script deacc.flux is executed to perform the de-accumulation of fluxes and copy the resulting files to the scratch filesystem.

The de-accumulation of precipitation (LSP, CP) data, is based on the following methodology:

a. Accumulated values for 3/6/9/... UTC are just divided by the number of hours, namely 3.
b. Accordingly, precipitation at X UTC is the mean precipitation per hour from X-3 to X UTC

For the de-accumulation of the other flux quantities the following strategy is used:

Accumulated values are divided by the number of hours and also interpolated to 3/6/9... UTC (for a three-hourly first guess with evaluation time X UTC, a flux value has been accumulated from X-3 UTC to X UTC and is therefore valid for X-1.5 UTC)

Accordingly, radiation (X UTC) is the radiation valid for X UTC (three-hourly average)

3) Retrieval of other meteorological fields and calculation of the vertical velocity component d(eta)/dt

These tasks are performed by the routines found at any of the directories: job_era, job_pre99, job_y2k, again according to the time period of the required GRIB data. Here the CONTROL file has to be edited, providing information like beginning date, ending date, time interval, number of levels to extract, scratch directory for (optional) flux data input, ecfile directory for output.

Afterwards, the FORTRAN sources are compiled **(f77 *.f $EMOSLIB).**

The execution of the resulting binary creates a batch script, which sends to the batch queue of the Mars archive and executes the routine CONVERT_PRE, which:

- Reads all ECMWF model data.
- Makes the wind fields 3D mass consistent, by calculating the vertical velocity (gridded values) component in the ECMWF eta vertical co-ordinate system (d(eta)/dt) applying the equation of continuity.
- Writes out all meteorological data including d(eta)/dt

- Saves the resulting output files (ECMWF GRIB format) in the user-specified directory of the EC File System.

3.5.2 Preparation of ECMWF GRIB data after July 2004

A number of changes to the way the meteorological data (ecmwf grib format)is retrieved from the computing systems of ECMWF occurred in July 2004. A summary of the more major changes include:

- New ECgate server
- ECgate1 system was switched off at the end of July 2004.
- ECbatch and ECcopy commands were terminated with the decommissioning of ECgate1.
- The new system (ECgate , an IBM p690 running AIX 5.2, 16x1.3GHz Power 4 CPUs and 32GByte Memory) uses the system LoadLeveler, rather than the previous NQS ("qsub"). LoadLeveler is a tool developed by IBM for managing serial and parallel jobs over a cluster of servers.
- All user scripts, using qsub commands required changing to use LoadLeveler instead.

As the original meteorological data extraction routines (Wotawa et *al, 1995*) were not compatible with the new ECMWF ECgate server systems, we developed new libraries for this research, based on the methodology and the source code of the original pre-processing routines developed by Wotawa, *et al.*

The meteorological data extraction routines were developed in June 2004, as a maintenance version of the discontinued extraction routines and methodology of Gerhard Wotawa, *et al.* 1995 according to specifications and the standards of the LoadLeveler system adopted by ECMWF's ECgate server after July 2004. The source code of the routines can be found in Appendices of this thesis and they became available for free download at our website:

http://www.gre.ac.uk/~ps60/ATTMA/downloads.htm

3.5.3 The revised GRIB data Preparation routines by G. Wotawa, *et al*

One month after our data retrieval tools were ported to the new ECgate system, Gerhard Wotawa, *et al* developed and made available a revised version of their data retrieval routines. The main characteristic of the revised routines was their compatibility with the ECMWF's LoadLeveler system. The new routines were (developed entirely from the beginning and were not based on the old routines. Also, they provided various improvements, including retrieval of the new early-cut-off ECMWF analysis data as well as operational and delayed-cutoff and data for higher than 1 degree resolution data (ECMWF data code: T511 instead of the old T319). The suggested early-cut-off analyses are actually operational data, used to get the forecast started quicker for a more timely delivery of forecast products than was the case before and with the delayed-cut-off analysis data.

The revised routines are a great improvement for the FLEXPART community, not only because of the compatibility to the new ECgate system and the revised methodology, but also because they offer a higher level of automation and users can retrieve and pre-process data by editing and running only one script. However the source code offered less configuration options such as the discontinued option for grid's spatial configuration: The new script is coded for the retrieval of global meteorological fields, which for a grid resolution of 0.5 degrees latitude/longitude results to almost 1.4 Gbytes of storage memory, whereas ECMWF users are allocated only 1 Gbyte of memory per data retrieval process (as of 2006). Therefore the updated Wotawa's scripts, data can be used for the retrieval of data with resolution of 1 degree. This issue has been discussed at the FLEXPART users newsletter. Hence for finer resolution of lower resolutions we continued to use our own ported routines, which later had some improvements imported from the new routines (including retrieval of short cut-off synoptic data, instead of operational data).

3.5.3 Final data retrieval, storage and preparation

The resulting GRIdded Binary (GRIB) encoded files (each for every different time interval) have to be transferred to the local workstation by using the ftp utility. However, the transfer of GRIB encoded files via ftp can lead to corrupted data, in which case the files have to be downloaded again. Alternatively ECMWF offers ftp tools for the safe transfer of GRIB encoded files.

It is recommended that all GRIB data files should be stored in a separate directory. In the same directory an ASCII file should be created, containing a list of the available weather files for each time period.

3.6 FLEXPART's Output and Post processing.

Several types of data can be output from FLEXPART's model run:

- Concentration levels (files grid_conc in unit 10^{-12} kg/m³);
- Mixing ratios (grid_pptv in Ad (files grid_conc* in unit ppt by volume);
- Residence times (in backward simulations, files grid time, unit seconds). All the above files are in binary (sparse matrix) format and contain traditional information about the uncertainties (standard deviation divided by mean concentration of all particle class) and the dry and wet depositions (unit 10^{-12} kg/m²).
- Plume trajectories. The sparse matrix binary format of the simulation's results does not allow for direct view and analysis of the output. Hence the files need decoding or conversion to "readable" formats such as the ASCII text or visualised graphs and plots. Plots and graphs out of the model's output can be created with the assistance of the NCAR Graphics package.

3.6.1 Visualisation of FLEXPART's output, using the NCAR Graphics package

The NCAR Graphics, developed by the US National Center for Atmospheric Research (NCAR), is a UNIX package, consisting of several Fortran/C utilities for drawing contours, maps, vectors, streamlines, weather maps, surfaces, histograms, X/Y plots, annotations, and more. The latest version 4.3.0 of NCAR Graphics is available for free with its source code or as binary files from the official web site of NCAR.

Some Fortran routines that employ the NCARG libraries and enable the conversion of FLEXPART's binary output to ncarg metafile, are available on A. Stohl's web site.

To plot FLEXPART results using the NCARG package, the source code of NCAR Graphics package was downloaded from the NCAR website (http://www.ncar.ucar.edu/ncar/). Alternatively the binary package of the same NCARG version can be used.

A Fortran 77 and an ANSI C compiler should be installed for a successful installation of NCARG and the GNU GCC and G77 were used for our installation. Also, the following X11 libraries are required, along with their associated include files:

X11, Xaw, Xext, Xm, Xmu, Xt

After the download and extraction, the source code was conFigured, compiled and installed, according to the installation instructions of the package.

The routines **FLEXPARTplot_vertsect.f** enable a vertical section plot (longitude or latitude) of the output between two predefined points.

After the extraction of the routines, all configuration files were edited to set the correct path to the directory where the output of FLEXPART is stored. Then the routines were compiled using the command **ncargf77** of the NCARG package, a script that invokes the FORTRAN 77 compiler/linker (in our case g77), with the proper NCAR graphics libraries.

Once the routines are executed, the NCARG metafile output can be converted into postscript (*.ps), by executing the command:

ctrans -d ps.color -f font12 gmeta > name_of_the_output.ps

The postscript file can be further converted into Portable Document Format (PDF) format, by applying the UNIX command ps2pdf, or into the Graphic Interchange Format (GIF) by using the command convert of the ImageMagick, am open source image manipuilation package which accommodates most UNIX operating systems.

3.6.2 Development of a postscript plotting library for FLEXPART

For better comparisons between the two models, a plotting facility had to be developed for FLEXPART aiming to provide an output result comparable with the postscript output of HYSPLIT. The facility was developed under the standards of Fortran 90 and using the libraries: "PSPLOT, PostScript for Technical Drawings - A free Fortran-callable PostScript Plotting Library". PSPLOT was developed by Kevin E. Kohler and its code is freely available as an open source project on the website: http://www.nova.edu/ocean/psplot.html.

The new libraries enable plotting of Lagrangian particle positions or pollutant concentration along a vertical or horizontal axis.

3.6.4 Development of a Graphical Unit Interface (GUI) for LPDM FLEXPART

The current version of FLEXPART is a console-mode application and the configuration process is performed by editing a series of ASCII text files. For a complete FLEXPART simulation, more than 15 text files have to be adjusted to the model's parameters, during both the FLEXPART's setup and the retrieval of GRIB weather data, remotely at the ECMWF's file systems.

Integrating a Graphical Unit Interface (GUI) to FLEXPART would make the use of FLEXPART more convenient, as it would benefit from the advantages that GUIs can offer, such as the ease of use and learning, the standardised interface, the ability to group and display relevant information and to perform multiple edits. A Graphical Unit Interface (GUI) based widget, with the proposed name "XFLEXPART", is under development as part of this research (figure 3.7), aiming to improve the configuration, control and monitoring of FLEXPART's operation. The interface will be available for download from the project's website and will be licensed under the GPL licence. XFLEXPART will be a controller and configuration tool for FLEXPART's core, so it i's applicable even in precompiled FLEXPART installations. It is developed in C++ programming language, using the wxWindows GUI framework, an open-source (LGPL licence) class library that allows the creation of graphical C++ programs on a range of different platforms (e.g. Windows/Unix/Mac). By linking with the appropriate wxWindows library of the platform (e.g. Windows/Unix/Mac) and the current compiler (almost any C++ compiler), XFLEXPART will adopt the look and feel of the platform's interface.

3.6.5 Availability issues

The availability of FLEXPART's source code and the right to amend it, enables unlimited customisations and improvements to the code. It also allows portability to new hardware, the easy adaptation to changing conditions/scientific knowledge, and reaching an advance level of understanding of how the methodology works. FLEXPART can be easily introduced to any project and be adapted to cooperate with existing software.

Furthermore, the right to redistribute modifications and improvements made to the code, and later to reintroduce further modifications made by other users, ensures that any improvements made by users can be available to everyone. And since users/groups can decide about the direction of their individual improvements, the overall direction of progress is determined by the collective desires of the user community.

In addition, the open source nature of the code makes FLEXPART a completely independent and self-regulated project. Even if the main authors (A. Stohl, *et al*) decide to discontinue the maintenance and improvement of FLEXPART (a common concern with proprietary software), the community may continue supporting and improving the code without any practical/legal limitation.

Moreover the above described benefits of FLEXPART's source openness, make it an attractive project for becoming a permitted, adopted and recommended dispersion model for governmental use in the European Community and worldwide.

All the above described benefits may attract more developers/users, which in turn helps to further increase the quality of the code, and to improve its methodology.

The disadvantages of FLEXPART, deriving from its community based nature are:

- Support issues. As in most open source projects there is no obligation by the developing group to provide official support to FLEXPART's user groups. However, in this research enthusiastic and varied unofficial support is generally offered by FLEXPART's developers and user community through email communication.

- Difficulty of use. Like most open source packages, FLEXPART aims at developers and users with high degree of technical expertise, for many of which it is accepted that ordinary operation of the model will involve creation of configuration files, writing scripts, or editing and recompiling the source code.

- Lack of capabilities and features. By depending exclusively on the user community, less features and capabilities are introduced to the reference version, compared to alternative proprietary models such as HYSPLIT (such as input/output facilities), where the authors can invest more effort as the model remains their property.

- Generally higher operational cost.Although FLEXPART is free and can be ported to any operating systems including open source ones (LINUX) it requires a much higher degree of technical expertise to operate and maintain, and as a consequence it may end up having higher operational cost.

3.6.5.1 FLEXPART's licence

Although it is generally considered as an open source project, FLEXPART is not currently covered by any well known official licence (e.g. approved by the Open

Source Initiative or other International Organisation). Alternatively the user's rights and obligations are defined by a note published by the authors on the project's website site:

"Access and use of FLEXPART shall impose the following obligations on the user. The user is granted the right, without any fee or cost, to use, copy, modify, alter, enhance and distribute FLEXPART, and any derivative works thereof, and its supporting documentation for any purpose whatsoever, except commercial sales, provided that this entire notice appears in all copies of the software, derivative works and supporting documentation.

This software is provided by the University of Munich "as is" and any express or implied warranties, including but not limited to, the implied warranties of merchantability and fitness for a particular purpose are disclaimed. In no event shall the University of Munich be liable for any special, indirect or consequential damages or any damages whatsoever, including but not limited to claims associated with the loss of data or profits, which may result from an action in contract, negligence or other tortious claim that arises out of or in connection with the access, use or performance of FLEXPART."

In general, following a well known and internationally approved license schemes offers various advantages including (Deutsch, 1996):

1. Guaranteeing particular basic freedoms (redistribution, modification, use) to the users.

2. Ensuring particular conditions imposed by the authors (for example citation of the author in derived works).

3. Guaranteeing that derived works are also open source software which is an important issue

4. Determining compatibility with proprietary software or other open source licensed cde. Prospective FLEXPART users/developers can easily determine whether portions of code from other projects can be inserted into FLEXPART (or the opposite). It is usually impossible to mix code not covered by any licensing scheme with licensed code (open source or propriety) in the same piece of software.

Furthermore all the above advantages are translated into probability of more developers being attracted to the project willing to invest effort and code on the project. Additionally an approved license scheme is the only way for FLEXPART to become a standard dispersion modelling method, approved and recommended for governmental use.

Therefore, adoption of an approved (open source) license scheme is highly recommended for the development team of FLEXPART.

3.6.5.2 The FLEXPART user community

FLEXPART is currently (2005) used by a relatively small community of 25 user groups (Stohl *et al*, 2005), therefore it does not take the maximum advantage of the above described benefits that community based projects usually offer. In addition, direct communication and collaboration between different FLEXPART's users has proved to be ineffective as users tend to put a large amount of effort into preparing the software and creating basic input/output facilities, spending effort in areas that in most cases have already been explored by other users.

Some the identified explanations for the small and ineffective user community include:

1. FLEXPART only aims at a limited user audience, due to reasons such as the limited application of LPD models, the UNIX character of FLEXPART having high technical expertise requirements etc.

2. Little attention has been paid by the authors to promote collaboration between users of FLEXPART, for example by the formation of an online user forum, where FLEXPART users could cooperate and easily exchange support and useful code. Cooperation between users was promoted by the authors only recently (since version 6.0), with the addition of a user list on FLEXPART's website. However this list is difficult to maintain and soon became outdated.

3. FLEXPART's source code is not licensed under an approved licence scheme (Open Source Initiative (OSI) or other organisation) making the project less attractive to new users and possible investors.

INSTITUTION	CONTACT	INPUT DATA USED	NATURE OF WORK
Equipe physico-chimie de la topophere (PCT), Service d'aeronomie, France	Augustin Colette	ECMWF ERA 40	Pollution transport
Laboratoire de Meteorologie Dynamique, France	Bernard Legras	ECMWF , Meso-NH,	Extensive recoding for stratospheric research, version called TRACZILLA. Fortran 90, namelist input, dynamic allocation, direct interpolation from model levels, etc. Meso-NH interface available and LMD-Z interface in development
French Atomic Energy Authority, France	Philippe Heinrich	MM5	WRF interface in development
Laboratoire de Physique de l'Atmosphere, Universite de La Reunion, France	Jimmy Leclaire de Bellevue	-	Stratosphere-troposphere in the vicinity of tropical cyclones and deep convective events
Radiological Safety Division, Indira Gandhi Centre for Atomic Research, India	C.Venkata Srinivas	MM5	-
Global Change Impact Studies Centre Islamabad, Pakistan	Rehan Anis and Shoiab Raza	ECMWF	Pollution transport
Instituto Nacional de Meteorologia, Spain	Jose Ardao-Berdejo	ECMWF	Operational FLEXPART runs
Meteo Swiss, Switzerland	Pirmin Kaufmann , Peter Roth, Thomas Egli	aLMo regional model,ECMWF	aLMo test installation, ECMWF version used operationally for emergency response applications
Institute for Atmospheric and Climate Science, ETH Zurich, Switzerland	Michael Sprenger and Junbo Cui	ECMW	Ozone trend over Europe and its link to atmospheric transport
University of East Anglia, U.K.	Kate Preston	ECMWF	-
NOAA Aeronomy Laboratory, USA	Owen Cooper	GFS	Pollution transport, stratospheric intrusions
Michigan Tech University, USA	Chris Owen and Drew Snauffer	ECMWF	-
Massachusetts Institute of Technology, USA	Benjamin de Foy	MM5	Mexico City Project, various changes to use MM5 PBL parameters, conversion of output to NARSTO-DES and NetCDF formats
Pacific Northwest National Laboratory, USA	Jerome Fast	WRF	source-receptor relationships, pollutant transport

Table 3.3 User groups of FLEXPART community and model's useage

Chapter 4

4. Model testing, hypothetical case studies

This chapter describes the simulation of a hypothetical pollutant release scenario. We compare weather models, predicted air concentrations and ground deposition in forward time and inverse mode in the relevant area defined as the Trans-Manche region. A hypothetical accident at the location of Dunkerque assumes a timed release of a gaseous pollutant. The calculations were performed using both LPD models HYSPLIT and FLEXPART and real weather situations, chosen at random. In order to compare the models and evaluate their performance and reliability in simulating the long-term dynamics of atmospheric pollutants, a sensitivity analysis with regard to both model formulation and input data has been carried out. Models have been driven by same input fields, and in particular:

- for the meteorological fields the MM5 mesoscale weather model and/or the ECMWF were used

Comparing model outputs obtained with the same input fields and the same configuration (as for example emissions and horizontal and vertical resolution) has allowed to highlight differences only due to models response. In particular, three main aspects were investigated:

a) prevailing weather conditions
b) horizontal model grid resolution and
c) horizontal resolution of input meteorology fields.

4.1.2 Model setup

In both setups a total number of 20,000 particles were tracked. Meteorological data of the European region was retrieved from the ECMWF Mars archive, for the period between 01-08-2002 and 04-08-2002, with a horizontal resolution of 0.5 degrees latitude/longitude, 61 vertical levels and 3 hours temporal resolution. The solution grid was conFigured equal to input data resolution grid (0.5 degrees).

A constant 1 hour release was assumed, starting at 15:00 on the 2nd of August 2002. Gaussian dispersion coefficients have been assumed in both models and short range dispersion mode was adopted for HYSPLIT (Horizontal and vertical velocity variances).

4.1.3 Results and discussion

The results of the simulation are shown in Figures 4.2a, 4.2b and 4.3 in the form of Horizontal (figure 4.2a, 4.2b) and vertical particle distribution (figure 4.3). Although the general plume trajectory is similar, the diffusion spread predicted by the two codes is very dissimilar. Differences are more apparent in the vertical plot of particle distribution highlighting the different method of treating the vertical component of wind velocities in the two models. Both models predict the plume returning to France at high altitude over the Bay of Biscay. Due to the different treatment of vertical motion, particles in HYSPLIT tend to accumulate behind frontal zones (figure 4.3). Detailed spatial and temporal meteorological data are critical to an accurate simulation. The data used for this example (horizontal resolution of 0.5° latitude/longitude) may have been insufficient for short-range applications.

4.2.2 Model Setup

In previous simulation (Case 1) FLEXPART and HYSPLIT were compared for this particular case, showing similarities but also substantial differences. This time, two HYSPLIT model runs were conFigured to simulate the release of SO_2, constant for duration of 1 hour, at 15:00 on the 2nd of August 2002. The model runs used the same configuration but different weather domain input data, provided by the MM5 model:

1. Three meteorological domains having resolutions of 36 km, 12 km and 4km, (domains D01, D02 and D03 in Figure 4.4).
2. A single weather domain of 36km (Domain 1 in Figure 4.4).

MM5 was initiated by a 6-hour ECMWF analysis data (as a boundary condition) and run to output weather data for the period 02-05 August 2002, with 1-hour temporal resolution and for the 3 domains shown in Figure 4.4. Table 4.1 summarises the MM5 model configuration settings, used in this case study.

Domain	Tree grids
Grid spacing	36,12 and 4-km horizontal grid spacing, with 60 verical layers.
Time step	100s
Initialization	ECMWF model, 2.5 grid
PBL	Blackadar
Soil model	multi-layer (ISOIL=1)
Nesting	Two-way, time-dependent
Output	Model fields saved every 1h
Timing	36h forecast requires approximately 6-7 hours elapsed time

Table 4.1 Configuration of mesoscale weather model MM5

Chapter 5

5. Real life case studies

In this chapter the modelling system was used to perform transport and dispersion calculations based on real meteorological situations and observed pollutant episodes, and the results were compared to historical data from ground level monitoring stations or satellite instruments.

Again, a sensitivity analysis was carried out with regard to both model formulations and input data. Models have been driven by same input fields, and in particular:

- the concentrations over all the domain were provided by the EXPER/PF database in the form of hourly average concentrations measured at ground level monitoring stations;
- for the meteorological fields the MM5 mesoscale weather model or alternatively the ECMWF were used;
- the boundary concentrations to iniate the MM5 model were provided by ECMWF

Comparing model outputs obtained with the same input fields and the same configuration (as for example emissions and horizontal and vertical resolution) has allowed us to highlight differences only due to models response. In particular, three main aspects were investigated:

a) prevailing weather conditions
b) horizontal grid resolution and
c) horizontal resolution of meteorology fields.

In the first (receptor) model run, HYSPLIT and FLEXPART were set using the same input fields. 20000 particles are evenly released in the Trans-Manche area of Folkestone (rural) and for the period between 7:00 and 8:00 and on the 3rd of March 2000). For HYSPLIT the Gaussian-Particle diffusion mode is selected to reflect the only option available in FLEXPART.

To model the North African desert dust storm, (second test case) the PM10 dust emission algorithm described by Draxler *et al.* (2001) was employed, as it is incorporated in HYSPLIT versions 4.6 and 4.7. The algorithm initiates dust emissions, up to a maximum value of 1 mg/m^2s^1, when the friction velocity exceeds a specified threshold velocity and from grid-cells (in this case 409) with a desert land-use classification. The particles are then dispersed and transported using HYSPLIT.

In test case 3, the simulation of the volcanic eruption at Hekla was based on observation data from the Icelandic Meteorological Office (IMO). IMO reported that the eruption began at 18:17 on the 26th of February 2000. The weather radar images showed no sign of an ash plume until 18:20, but a plume at this site has to reach a height of 2 km before it is detected by the radar. On the next radar image at 18:25 the plume had reached a height of 11 km (Ragnar Stefánsson, IMO, 2000). To simulate the plume rise, a vertical line source was set, extending from the volcano summit to the eruption column top and 20,000 particles were released at Hekla (latitude 64.26° N , Longitude: 17.26° W) and dispersed using FLEXPART . HYSPLIT was not used for this simulation.

5.2.2 Model Setup

A series of forward and inverse simulations were performed using LPD model FLEXPART. Air quality data (PM10 concentrations) were retrieved (figure 12) and global meteorological fields datasets for the period between 13 to 20 March 2004 from the ECMWF Mars archive having horizontal resolution of 0.5 degrees lat/lon and 60 vertical levels. 200,000 notional particles were tracked for the area of Rochester (automated rural station), between 17:00 and 18:00 and on the 20rd of March 2000, when the peak concentration was recorded in Rochester.

A forward simulation of the eruption of Soufriere Hills, Montserrat volcano (Location: 16.7N, 62.2W, Elevation: 915 m) was run, based on observations from the Montserrat Volcano Observatory (http://www.mvo.ms) about the height of the plum. Then a column source was conFigured applyinmg the same methodology that was applied in the case study of Hekla volcanic eruption (paragraph 5.1). The results are illustrated in paragraph 5.2.3.

5.4.2 Model Setup

In order to model the dust emission caused by the Sahara desert storm, the revised (2002) dust emission model of R. Draxler *et al* (2005) is employed, as it is incorporated in the source code of HYSPLIT version 4.7. The emission module of HYSPLIT is based on the concept that threshold friction velocity is dependent on surface roughness. Surface roughness then is correlated with soil properties. The emission area specified by the user is divided into a matrix of points. Then those points are set as possible point sources in HYSPLIT, if they are located inside the area identified as desert in HYSPLIT's land-use internal database. A dust emission rate is computed from each cell of the resulting emission matrix, when the local wind velocity exceeds the threshold velocity for the soil characteristics of the emission cell (Westphal *et al*, 1987). Finally, the emitted material is dispersed and transported using the standard methodology of HYSPLIT (Draxler and Hess, 1998).

The recently revised methodology (Draxler., 2002) is expected to over-predict the air concentrations. Part of the over-prediction can be attributed to the model's sensitivity to the threshold friction velocity and the surface soil texture coefficient (the soil emission factor), as well to the difficulty in accurately representing these parameters in the model (Draxler *et al*, 2004). More information about the methodology of HYSPLIT is provided in chapter 3.

Meteorological data were retrieved from the ECMWF Mars archive, using our revised routines (see chapter 3.4.2) which were based on the original data preparation routines provided by G. Wotawa *(Wotawa et al, 1995)*. Dry depositions were switched off, in agreement with the methodology followed by Draxler R. (2002). The results are compared with pm10 concentration values measured at Finokalia, Crete and with satellite Aerosol Index data from the OMI instrument.

Then, LPD model FLEXPART is conFigured to simulate backwards in time the first dust event, observed at the Finokalia Station in Crete, on 24/02/06 at 14:30 (2,717 μg/cm³). 100,000 particles were released and FLEXPART model was set with a solution grid of 0.5 degrees to run for the period 23/02/2006 to 01/03/2006. The same synoptic data from the Mars archive of the ECMWF were used.

prediction is due to the background PM10 concentrations that contribute at the measured values.

The methodology could also easily have been applied to predict desert dust storms and the dispersion of dust, if forecast data was used instead of analysis data. The emission algorithm incorporated in HYSPLIT will automatically initiate particles whenever the wind speed exceeds the threshold value.

The output of FLEXPART's backward tracking, shown on the Figure 5.28 (left column) confirms the probability of the area of North Africa as the most possible source PM10 location for the first event in Crete (24/02/06). This agrees with HYSPLIT's forward simulation (figure 5.28, right).

5.4.4 Discussion

Several examples were presented of how Lagrangian transport and dispersion models can be used to simulate desert storms. Although the PM10 emission module of HYSPLIT over-predicted the first exceedences and under-predicted the PM10 concentrations during the second and third episodes it gives a valuable indication of the timing and the magnitude of the events, even though the dispersion was over long distance and considerable time span.

Part of the under-prediction is due to the background PM10 concentrations that contribute to the measured values. Over-predictions can be attributed to the resolution and the accuracy of HYSPLIT's land-use dataset, the model's sensitivity to the threshold friction velocity and surface soil texture coefficient, as well as to the difficulty in accurately representing these parameters in the model (Draxler R., 2004).

The quality of the simulation can be improved by an improved calibration of the threshold friction velocity and surface soil texture coefficients and by introduction of a more realistic and detailed land use dataset.

The methodology could also easily have been applied to predict the dispersion of the dust due, if forecast data was used instead of analysis data. The emission algorithm incorporated in HYSPLIT automatically initiates particles whenever the wind speed is sufficiently strong.

The on-line NASA TOMS and the new OMI Aerosol Index database can be a valuable resource of information in validating qualitatively the model's output results. However such validations cannot be quantified, as Aerosol Index data contain no information over the vertical distribution of dust and they can only be used to indicate of the dust plume's location.

Finally, there was a good agreement between the results of the two codes, namely FLEXPART's inverse mode based on receptor data and HYSPLIT's desert storm (forward) model.

Chapter 6

6. Conclusions

6.1 Overview

This research is concerned with methods of interpreting air quality monitoring data and in specific, with numerical modelling of atmospheric dispersion. The relationship between the composition of air and its origin is best interpreted by calculating the path of air masses backward in time from a monitoring site (Flesch *et al.*, 1995), (Seibert *et al.*, 2004).

The main objective of this research is to propose a modelling system, a methodology that can be applied to evaluate and interpret source-receptor relationships for atmospheric pollutants from several locations to geographical regions of Europe. For this purpose, various research tools were applied:
dispersion modelling, trajectory modelling, probability fields analysis of trajectory and dispersion modelling results.
It is generally agreed that output from air quality numerical models should be viewed as containing both reducible error and inherent uncertainty (e.g. (Fox *et al*, 1984; Fisher *et al*, 2001). Reducible error results from improper or inadequate input meteorological and air quality data and from inadequacies in the models (e.g. resolution of the solution grid). Inherent uncertainty results from the stochastic nature of the turbulent atmospheric motions that are responsible for transport and diffusion of released compounds.

The magnitude of the errors and uncertainty and hence the ultimate limit for using numerical modelling methods to calculate the sources from which an air quality measurement originates, can be investigated using Lagrangian Particle Dispersion models. The performance of Lagrangian Particle Dispersion models can also be used to investigate the accuracy of predictions of the future movement air pollutant clouds using the three-dimensional wind field and the integration of the velocity with respect to time to obtain the trajectory co-ordinates. Each trajectory is also affected by random movements associated with stochastic fluctuations caused by turbulence. There has been much discussion within the British dispersion modelling community regarding the possibility of using more than one model, to investigate the accuracy of predictions (Fisher et al, 2002).

This study describes a modelling system proposed for the ATMMA project which comprises of:

- Two trajectory and dispersion modelling systems as are incorporated in the Lagrangian Particle (LPD) models HYSPLIT (Draxler, 2001) and FLEXPART (Stohl *et al* 2005)
- A meso-scale weather model, namely MM5
- Input air quality data from the EXPE/PF, the U.K. Air Quality Archive, Kent Air Quality Network databases and Aerosol Index (satellite) data from TOMS and OMI.
- Input meteorological boundary conditions provided by the ECMWF
- A web interface where partners can request a model run available at www.gre.ac.uk/~ps60/attma

The modelling system was applied to a number of real cases and hypothetical scenarios. Principal considerations about the evaluation of mesoscale atmospheric dispersion models are presented, exploiting and commenting a concept which was developed in the framework of the Anglo-Franco Aerosol transport research project ATTMA. Some of the difficulties of the evaluation of the accuracy of the models are limitations and inaccuracies of the input data, such as the meteorological fields and emission data or air quality measurements. One of the major concerns of this research is to evaluate the applicability and reliability of the Lagrangian Particle models as tools for air quality assessments in European regions.

Specific experiences resulting from the application of the modelling system are described in chapter 6.3. It is expected that the most valuable applications will arise when there is a single major source, of uncertainty magnitude, some several hundred kilometres from the monitoring sites. Some significant differences in the way the models behave have been identified. Conclusions concerning the way in which the models should be used are drawn.

This work has been achieved in close collaboration with other European partners in the context of the ATTMA project and with other collaborations from the user community of FLEXPART LPD modelling system. Our contribution to both purposes and more generally to the research on the air quality modelling and output visualisation, is summarised in this concluding chapter and specifically in section 6.2. Perspectives of potential future investigations and developments are finally proposed in section 6.4, as a conclusion to this work.

6.2 Contribution

This research made a number of significant and original contributions in the area of numerical atmospheric dispersion modelling. Contributions described in this dissertation can be summarised as:

5. Proposed a atmospheric dispersion modelling system for estimating source-receptor relationships in the Trans-manche region, consisting of two Lagrangian models (FLEXPART and HYSPLIT), using a mesoscale weather model (MM5).

6. Modified the MM5 version of FLEXPART (version 3.1) (*Wotawa* refe to introduce updates and features of the ECMWF reference version (version 6.2), such as the new output file format and improvements in the handling of convective mixing.

7. Compared and evaluated the atmospheric transport between two widely used Lagrangian Particle Dispersion models, namely FLEXPART (*Stohl et al., 2005*) and HYSPLIT (*Draxler and Hess, 1997*) and explored their differences, features and limitations.

8. Developed various input/output facilities for community based Lagrangian Particle dispersion model FLEXPART and made them available to the FLEXPART community such as: a visualisation tool for FLEXPART's binary output and a converter to readable ASCII text files.

9. Demonstration with real world examples the sensitivity of these LPD models to input data, grid resolution, input data and configuration.

6.3 Concluding remarks about the ATTMA modelling system

6.3.1 Overview

This chapter demonstrates concluding remarks and specific experiences resulting from the application of the modelling system. In this research two Lagrangian Particle dispersion models, namely HYSPLIT and FLEXPART performed transport and dispersion calculations of real life pollution events and hypothetical scenarios, both based on realistic meteorological cases. The models were initiated with meteorological data from the MM5 mesoscale model and/or data boundary conditions derived from the ECMWF Mars archive. For the real examples, the simulation output was compared to historical data from ground level measurements or/and with data from satellite instruments, such as AI data from TOMS and OMI.

Differences between the results of the two models raise questions over the reliability of dispersion models and stress the need for more than one dispersion model in air quality simulations. Despite the differences, which are under investigation, the simulations give a useful indication of the location of possible sources of the pollution even over long distances and considerable time span.

6.3.2 Applicability

6.3.2.1 Overview

This study has shown, that Lagrangian particle models have been developed, validated and used intensively in recent years and are available for the assessment of air quality assessment due to releases of airborne pollutants within a wide range of situations (sources/distances). Depending on the physical contents, the Lagrangian particle models possesses a greater significance compared to Eulerian and Gaussian plume models and in general do not have the restrictions mentioned above.

The modelling system can support a wide range of simulations related to the long-range transport, dispersion, and deposition of pollutants. The applications can range from the need to respond to atmospheric emergencies, ranging in character from accidental radiological releases to the hazards presented to aircraft operations from volcanic ash eruptions, or routine air quality assessments such as those associated with emissions of anthropogenic pollutants. Simulation output results can vary from simple trajectories to more complex air concentration contour patterns. Calculations can be performed on archive or forecast meteorological data, or a combination of both.

However it should be kept in mind that LPD models require more measurement input data and accurate meteorological fields to be able to output reliable and realistic results. But as most of these data can be provided either by dispersion parameterisation, by the use of pre-processors or weather services, further investigations on the introduction of Lagrangian particle models for regulatory purposes in European Union can be strongly recommended.

Even though this study reveals a positive attitude to further investigations for the introduction of Lagrangian particle models into European regulations concerning source apportionment of sources responsible for air quality problems, it should be nevertheless kept in mind, that most of the advantages of this type of models only can be gained if a suitable meteorological fields are applied in combination. But introducing a new model chain, e.g. based on pre-processors with realistic pollutant release data on the emission site, meteorological fields from a CFD model and Lagrangian particle model, a new level for the assessment of air quality assessments can be realised.

There are some types of complicated scenarios where the proposed model has limited applicability. Both Lagrangian Models have difficulty resolving flow and dispersion through complex terrain such street canyons, hills and valleys and generally on scales not resolved by the meteorological model. HYSPLIT and the reference version of FLEXPART are intended to be used for neutrally buoyant releases and are not able to model gases whose density effects remain significant at downwind distances beyond about 1 km. However, they can be applied to heavy gas releases that are rapidly diluted. Brett Anderson (EPA, USA) is developing a version of FLEXPART that enables limited support for buoyancy.

The modelling system applies to gases and particles with low chemical reactivity. In situations where there are strong and/or non-linear chemical reactions, such as the photochemical reactions responsible for the formation of Ozone, the chemicals that are initially released may nearly disappear as several secondary chemicals are formed and the applied LPD models are not capable of treating these complicated reaction mechanisms

Situations where the proposed system is applicable	**Situations where the proposed system is NOT Applicable**
	complex terrain such street canyons, hills and valleys and generally on scales not resolved by

	the meteorological model
Neutrally buoyant releases	
Heavy gases that behave like neutrally buoyant gases due to rapid dilution	Heavy gases with significant prolonged density effects
gases and particles that have limited chemical reactivity	Transport & dispersion effects resulting from non-linear chemical reactions
	Plume rise resulting from chemical reactions that release significant amounts of heat

Table 6.2 Applicability of the Modelling System

Also, as a result of some chemical reactions, heat may be released or absorbed, and both Lagrangian models do not account for those effects. An example of a source with heat released as a result of chemical reactions is oleum, or fuming sulfuric acid, which reacts with atmospheric water vapor to form sulfuric acid vapor and mist, releasing significant amounts of heat. For large oleum releases, plume rise can be significant, so it is important to input good on-site visual observations of the plume rise.

6.3.2.2 Applicability of Aerosol Index (AI) data for validation of Atmospheric Dispersion problems

Draxler R. et al (2002, 2004) have demonstrated that Aerosol Index data can be valuable tools in validations of desert storm LPD model outputs, especially when ground-level measurements are not available. In this study we presented various examples of modelling desert dust storms with LPD models and used Aerosol Index data from TOMS and OMI for the validation of the LPD modelling output (paragraphs 4.2.3.2 and 4.6.3). Aerosol index data could be used to correct the position of the predicted plume and then re-initiate the simulation. However, AI data are only qualitative means of identifying the plume's location, as particle index is averaged in the vertical axis.

6.3.3 Evaluation and Reliability

The general method of evaluating the accuracy of output results from atmospheric dispersion models is usually quite straightforward: simulation output is compared against appropriate measurements and identified deviations are statistically analysed to quantify the model uncertainties. In spite of the simplicity of this method, there are considerable difficulties in terms of defining the evaluation procedure and the interpretation of the results. Deviations between simulation output and observations can originate many reasons such as the specific assumptions followed by the model and it's parameterisation, errors and inaccuracies in input meteorological data (lack of accurate emission data), uncertainties related to the stochastic nature of modelling atmospheric processes, uncertainties in observations, uncertainties in the representativeness of both observed and modelled data etc.

The qualitative comparisons of observed and predicted concentrations on case studies of chapter 4 indicate that both LPD models can generally yield reasonable good results. Fields of observed and predicted concentrations have a similar geometric form and concentrations values correspond in order of one magnitude.

However, reasons such as the lack of emission data make the accurate quantitative evaluation of models performance practically impossible, in both forward simulations, where the dispersion and transport of emitted material is simulated with positive time step and in inverse mode, where probability contours are generated from a receptor based backward simulation. Another reason for the difficulty in accessing the accuracy of models HYSPLIT and FLEXPART is the inappropriateness of the Trans-Manche region for such a research, because it is surrounded by many pollutant sources. Furthermore inverse simulations in this thesis showed that both models predicted the timing and magnitude of pollution events.

The following comments can be made regarding the reliability of the proposed modelling system:

1) **Sensitivity of the results to the horizontal resolution of the input meteorological fields**

   Due to inadequate diffusion methodology followed by the models, the position of particles is dependant to the grid resolution at the release point, with higher resolutions producing finer initial plumes. However the relation between the resolution of meteorological. data and the accuracy of output results is clear, meaning that increasing the resolution of input data the accuracy does not necessarily lead to an improved simulation output. The issue is highlighted in simulations with multiple nesting, when particles move across nested grids with different resolutions. Further research is needed to identify the most suitable grid resolution that provides the most realistic results, or

2) **Reliability issues due to the differences in the results between the two LPD models**

   Comparison of models HYSPLIT and FLEXPART shows that there are notable differences between the two formulations, although both models are driven by the same meteorological data. A major reason is the different method that the models use to handle the vertical motion of particles. Another reason is that FLEXPART has a scheme to parameterise convective mixing, while HYSPLIT does not. Differences depend on meteorological conditions and this will be investigated in further studies. This suggests that the identification of the sources of air pollutants may be rather uncertain, highlighting the need of using more than one models and making maximum use of the available air quality data.

3) Sensitivity to configuration options

Choices in the model configuration of both the Lagrangian Models and the preparation of meteorological data can highly affect the result. Examples include dispersion method in HYSPLIT (puff-particle)

The preparation of GRIB data within the Mars archive of ECMWF strongly affects the output results.

Options in HYSPLIT include the choice of the diffusion mode (normal – short range) and dispersion method (Top-Hat or Gaussian)

Options in FLEXPART include account for subgrid-scale convective transport, time constant for particle splitting and synchronisation interval.

Additional tests are needed to identify combinations of input options.

ı.3.4 Availability of the Model

'able 6.2 presents the availability of the major compontents of ATTMA modelling ystem. One of the most important differences between the LPD models FLEXPART nd HYSPLIT is that FLEXPART is an open source, community based project while IYSPLIT is freeware propriety software. Legally, the difference is that open source llows users to access the source code while freeware does not. However, ırganisationally, this leads to a big difference:

'here is no community and no development infrastructure around "freeware" as there s around open source software. Thus, while users can use freeware "as is," there is no eal way to improve upon it or obtain community support for it.

;omponent	Availability	Accessible at:
'LEXPART	Open – source code	http://zardoz.nilu.no/~andreas/flextra+flexpart.html
IYSPLIT	Freeware (binaries)	http://www.arl.noaa.gov/ready/hysplit4.html
ıM5	Open- source code	http://www.mmm.ucar.edu/mm5/
;CMWF Mars \rchive	User account required. Free met. data (up tp 0.25 degrees) for UK and non-commercial users	http://www.ecmwf.int
;CMWF 'ree data service ERA-40)	Low resolution data (2.5x2.5) for period (09/1957-08/2002)	http://data.ecmwf.int/data
;XPER/PF ıir quality latabase	Air quality data for the Trans-manche region available only to ATTMA members	http://82.127.4.221/convertionAnglais.php
JK Air Quality)ata and ;tatistics)atabase	Free data from over 1500 monitoring sites across the UK	http://www.airquality.co.uk
\erosol Index lata (Satellite)	Free AI data from Earth Probe TOMS and the Ozone Monitoring Instrument (OMI)	TOMS: http://jwocky.gsfc.nasa.gov/eptoms/ep.html OMI: http://aura.gsfc.nasa.gov/instruments/omi/index.html

Table 6.2 Availability of the major components of ATTMA modelling system

The open source nature of FLEXPART offers great advantages but also big disadvantages as they have been described in chapter 3.6.1. The main advantages experienced in this research are:

- Portability to any hardware. As long as cross-platform code is used, FLEXPART can be compiled in any compiler or hardware (i.e. Sun Solaris with f77 compiler, Linux with G77 compiler).

- Flexibility of FLEXPART's code to be adapted and introduced to this research project and right to the user to create a different version of FLEXPART and redistribute it. In this research we used the MM5 version of FLEXPART developed and kindly by Bret Anderson (Environment Protection Agency, USA) which was based on the MM5 modified version of G. Wotawa et al. Bret Andreson updated the Wotawa's MM5 version (reference to original Sohl's version: 3.1) to version 3.2 (as corresponds to A. Stohl's reference version using ECMWF data) to include flux planes, age spectra, on-line clustering, and Emanuel convective parametrisation. We further updated Anderson's version to import improvements from newer FLEXPART versions, such as the new output format and the updated Emanuel convective parametrisation. Furthermore, Caroline Forster (Norwegian Institute for Air Research, Norway) ported FLEXPART version 6 to use data from the GFS weather model (Kalnay and Kanamitsu, *1988*) and due to its open-source nature, FLEXPART was the first model ported to operate with data from the WRF Mesoscale Model (Janjic, , *et al 2004*) as Jerome Fast (Pacific Northwest National Laboratory, USA) developed a WRF version of FLEXPART optimised for mesoscale use. Interface for the ALADIN Numerical Weather Prediction Project has also been developed by Helfried

Scheifinger and Mathias Langer (Zentralanstalt fuer Meteorologie und Geodynamik, Austria)

- Opportunity for FLEXPART's code maturity and excellence: As more people have access to the source code, more bugs were identified and related solutions were applied in newer versions. Furthermore, FLEXPART's methodology is challenged and improvements were offered by users. An example is the complete revision of the original convection scheme (Seibert *et al*, 2001).

- Independence: As long as there is available interest in FLEXPART by user groups, it will continue to evolve and be maintained by the community, without any practical or legal issues, even if the initial authors decide to change FLEXPART's objectives or abandon its development.

- The open nature of FLEXPART makes it an ideal tool for academic purposes and potentially a dispersion modelling standard for use by authorities and governmental organisations.

On the other hand the propriety nature of HYSPLIT offers the following advantages over FLEXPART:

- More capabilities and features: As the code has legal ownership, more effort can be invested by the authors/owners for the addition of features and make it a complete application.

- Easy of use. There is no requirement for the user to have a high degree of technical expertise, compared to open source projects that in most cases require editing configuration files, writing scripts, or editing and recompiling the source code.

- Generally lower operational cost. As it requires a much lower degree of technical expertise compared to FLEXPART.

From the above, it can be suggested that HYSPLIT is more suitable for user groups that require an easy to use and easy to learn model, with all pre-processing and post-processing tools included in the package and with low requirements in terms of technical expertise (e.g. no need for scripting, coding or compiling). On the other hand, adaptation of FLEXPART is more suitable for large scale research projects, for research groups with high technical expertise and any case where flexibility, independence and access to the source code are of high importance.

6.4 Future Direction

In this study the ATTMA modelling system was tested using the primary capabilities of the two LPD models including:

- Forward / backward dispersion
- Forward / backward trajectories
- Receptor concentrations

Backward simulations have been more thoroughly tested than forward simulations. Additional testing is needed to thoroughly test other capabilities of the code including emission variation files, removal processes, uncertainties, age spectra, mass fluxes, and the simulation of atmospheric trace gases such as stratospheric ozone.

The system should work for most forward/backward trajectory and dispersion applications. Nevertheless, our experience with the code has identified several issues that should be addressed for long-term use of the system including:

1) Graphical Unit Interface (GUI) for FLEXPART.

2) Turbulence: Turbulent wind components are currently calculated using the parameterisations described in Stohl et al. (2005). It would be useful to have the option of making the turbulent components consistent with turbulence kinetic energy predicted by the weather models.

2) Comprehensive testing of Input Options: Additional tests are needed to identify combinations of input options in the LPD models and the MM5 Mesoscale weather model.

3) Near-surface meteorological quantities: FLEXPART computes 10-m winds and 2-m temperature based on the coarse resolution output from the ECMWF model. An option needs to be added to deactivate this when the Mesoscale model simulation (MM5) has fine vertical grid spacing near the surface.

4) A dust emission module would be useful for FLEXPART to enable forward simulations of desert dust storm, similar to the module incorporated in HYSPLIT

5) Further development of post-processing tools for both HYSPLIT and FLEXPART

6) Development of pre-processing tools to enable the use of meteorological data from various sources such as the UK Unified model, Phoenix CFD.

7) Chemistry model. Current versions of FLEXPART and HYSPLIT only simulate the transport and dispersion of non-reactive aerosols and gasseous pollutants. It would be useful to couple those models with a chemistry model, to enable the simulation of phenomena such as photochemical reactions responsible for the Ozone formation.

Appendices

Appendix 1: Copy of the Presented Paper on the 5th International conference on Urban Air Quality

Plainiotis S., Pericleous K.A., Fisher B.E.A, Shier L., 2005: Modelling high particulate matter and ozone episodes in the Trans-Manche region. Proceedings of the 5th International conference on Urban Air Quality, March 2005

MODELLING HIGH PARTICULATE MATTER AND OZONE EPISODES IN THE TRANS-MANCHE REGION

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ABSTRACT

The ATTMA "Aerosol Transport in the Trans-Manche Atmosphere" project investigates the transportation and dispersion of air pollutants across the English Channel, in collaboration with local authorities and other Universities in Southern England and Northern France. Two widely applied Lagrangian Particle Dispersion (LPD) models are used and compared. In many episodes the possible source of air pollution is traced outside the region of interest, hence long range, trans-continental transport is also investigated.

1. INTRODUCTION

There is concern in the Cross-Channel region of Nord-Pas-de-Calais (North-West France) and Kent (South-East Great Britain), regarding the detected extent of atmospheric pollution from emitted gaseous (VOC, NOx, SO2) and particulate substances in the atmosphere. In particular, the air quality of the Cross-Channel or "Trans-Manche" region is highly affected by the heavily industrial area of Dunkerque, France, in addition transportation sources in Kent, are posing threats to the environment and human health.
Lagrangian Particle Dispersion (LPD) models are often used to numerically simulate the dispersion of a passive tracer in the planetary boundary layer by calculating the Lagrangian trajectories of thousands of notional particles. The project investigated the use of two widely used particle dispersion models: the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) Model and the model FLEXPART. In both models forward tracking and inverse (or receptor-based) modes are possible.
Certain distinct pollution episodes have been selected from the Franco-Belgian database EXPER/PF and also from UK monitoring stations and their likely trajectories were predicted using prevailing weather data. Global meteorological datasets were downloaded from the ECMWF MARS archive. Part of the difficulty in identifying pollution sources arises because much of the pollution originates outside the monitoring area. For example heightened particulate concentrations can originate from dust storms in the Sahara or volcanic activities in Iceland or the Caribbean.
One of the principal aims of this project is to evaluate, improve and compare existing models and hence propose the most suitable methodology for each situation. This paper contains a very brief description of the methodology followed in this project, in particular the numerical models within FLEXPART (Version 5.0) which was kindly provided in source code form. This is followed by a description of the input data used and examples of typical pollution episodes.

2. DATA AND METHODS

A number of requirements must be met for credible air pollution modelling. The mathematical models discussed in this paper use input data (meteorological, emissions, land use etc.) to predict pollutant concentration, or to identify pollution sources.

2.1 Particle Dispersion Model "FLEXPART 5.0"

FLEXPART is a Lagrangian Particle Dispersion (LPD) model, developed by Andreas Stohl (University of Munich) and is widely used to calculate the transport and dispersion of non-reactive pollutants in the atmosphere. It is an open source code (Fortran 77) project, freely available for scientific use; hence it can build a good base for the needs of our simulations. Psplot, a free PostScript plotting FORTRAN library, developed by the NSU Oceanographic Centre, USA, was adapted for use with this code.
FLEXPART has been validated with measurement data from large-scale tracer experiments, during case-studies of stratospheric intrusions and has performed well in comparison with other similar models [Stohl *et al*]. In this paper FLEXPART will be evaluated with other comparable models such as the HYSPLIT model.

2.2 The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) V.4.7 Model.

HYSPLIT is the result of cooperation between the National Oceanic and Atmospheric Administration (NOAA) and Australia's Bureau of Meteorology. The dispersion of a pollutant within HYSPLIT is calculated by assuming either a Gaussian or a top-hat distribution within a puff or from a fixed particle position [Draxler, *et al*]. HYSPLIT is coded in Fortran-90 and incorporates its own Graphical User Interface and several tools to analyse and post-process the output of air concentration, trajectory, or precipitation simulations.

2.3 Meteorological Input data

Aerosol dispersion models generally use meteorological fields and emission inventories as input, and calculate time-dependent concentration fields as output, using suitable models for the relevant processes such as advective or convective transport, mixing by diffusion, and mass transfer through dry or wet deposition, or chemical reaction. Both models, in this project, are driven by meteorological data from the Mars archive of the European Centre for Medium-Range Weather Forecasts (ECMWF), encoded under the WMO FM-92 GRIdded Binary (GRIB) format. The data sets within ECMWF are based on quantities analysed or computed within the re-analysis data assimilation schemes or from forecasts based on these analyses. An authorised user account is granted for this project, which allows for access to the ECMWF computing facilities. The data is retrieved and pre-processed using the ECMWF computing facilities and can have a horizontal resolution of up to 0.5 degrees, 60 vertical levels and temporal resolution of 3 hours.

2.4 Atmospheric Pollution Data

The "EXPER/PF database" project
EXPER/PF (Exposure of communities living in the centre of the Euro region to polluting atmospheric particles: the case of fine particulate matter) is a project under the framework of the INTERREG III programme. It combines air quality networks regular measurements and PM maps, derived from satellite data (MERIS, SeaWiFS).

The UK Air Quality Information Archive
The Air Quality Data and Statistics Database offers the retrieval of statistics related to Air Quality in the UK, from the present back to 1960, from the monitoring networks operated on behalf of the Department for Environment Food and Rural Affairs, UK (DEFRA) and the devolved administrations.

The Kent Air Quality Monitoring Network
The Kent Air Quality Monitoring Network provides a co-ordinated means of monitoring air quality throughout the county. Information on levels of 6 pollutants (carbon monoxide, ozone, nitrogen dioxide, coarse particles, fine particles and sulphur dioxide) is distributed every day.

3. BRIEF MATHEMATICAL DESCRIPTION OF FLEXPART

LPD FLEXPART takes into account wind velocity variances and Lagrangian autocorrelations to determine appropriate diffusion coefficients. Particle dispersion is then modelled via the Langevin equation as derived by Thomson for inhomogeneous and Gaussian turbulence under non stationary conditions [Stohl *et al*]. Turbulence statistics are obtained by using the Hanna scheme with some modifications taken from Ryall and Maryon for convective conditions.

FLEXPART, like most Lagrangian particle dispersion models (LPDM), assumes a Markov process to calculate the velocity and position of particles [Stohl *et al*]. Gaussian turbulence is assumed, which is accurate only in stable and neutral conditions. FLEXPART implements a parameterisation of convective mixing. As the meteorological fields from the European Centre for Medium-Range Weather Forecasts (ECMWF) provide no direct information on convection, a convection scheme is employed to provide the convective redistribution of air from the resolved scale fields of temperature and humidity.

In addition, wet deposition can be calculated in FLEXPART using scavenging coefficients. Backward simulations can be performed by applying the same methodology and numerical model, but with the particle trajectories being integrated backwards in time, so using a negative time step. The methodology in FLEXPART is based on mass mixing ratios, rather than mass consistent concentrations and these particles are only sources, used for the determination of the trajectories and linear-source receptor relationships for the transport of atmospheric trace substances.

Because HYSPLIT is better known, it is not described in detail. As it uses a prescribed concentration

5. CONCLUSIONS

In this study, we present parallel simulations of typical pollution episodes. We have used two Lagrangian methods for this purpose as incorporated in the codes of FLEXPART and HYSPLIT. Particles are released in prescribed source regions, and transported with the mean and a stochastic turbulent velocity field. Particles can then be tracked in forward and backward mode.
The output of the simulations shows that there are notable differences between the two formulations, although both models use the same meteorological data and source input.
This means that the identification of the primary emissions during air pollution episodes may be rather uncertain. Hence, there is the need to make maximum use of the available data.
Despite the differences which are under investigation, the simulations give a useful indication of the location of possible sources of the pollution even over long distances and considerable time span.

6. REFERENCES

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Appendix 2: Copy of the Presented Paper on the 16th IASTED International Conference on Modelling and Simulation

Plainiotis S., Pericleous K.A., Fisher B.E.A., and Shier L., 2005: Forward and Inverse Transport of Particulate Matter and Gaseous Pollutants Affecting the Region Bordering the English Channel, Proceedings of the 16th IASTED International Conference on Modelling and Simulation (MS-2005), pp.164-169, Acta Press pp.459-090

FORWARD AND INVERSE TRANSPORT OF PARTICULATE MATTER AND GASEOUS POLLUTANTS AFFECTING THE REGION BORDERING THE ENGLISH CHANNEL

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ABSTRACT

The ATTMA "Aerosol Transport in the Trans-Manche Atmosphere" project investigates the transportation and dispersion of air pollutants across the English Channel, in collaboration with local authorities and other Universities in Southern England and Northern France. The research is concerned with both forward and inverse (receptor based) tracking. Two alternative dispersion simulation methods are used: (a) Lagrangian Particle Dispersion (LPD) models, (b) Eulerian Finite Volume type models. This paper is concerned with part (a), the simulations based on LPD models. Two widely applied LPD models are used and compared. Since in many observed episodes the source of pollution is traced outside the region of interest, long range, trans-continental transport is also investigated.

KEY WORDS

Atmospheric Pollution, Lagrangian, Modelling, Air quality

1. Introduction

There is concern in the Cross-Channel region of Nord-Pas-de-Calais (North-West France) and Kent (South-East Great Britain), regarding the detected extent of atmospheric pollution from emitted gaseous (VOC, NOx, SO2) and particulate substances in the atmosphere. In particular, the air quality of the Cross-Channel or "Trans-Manche" region is highly affected by the heavily industrialised area of Dunkerque, France, in addition to considerable emissions from transportation sources in the port regions of Kent (UK), posing threats to the environment and human heath.

Lagrangian Particle Dispersion (LPD) models are often used to numerically simulate the dispersion of a passive tracer in the planetary boundary layer by calculating the Lagrangian trajectories of thousands of notional particles. The project investigated the use of two widely used particle dispersion models, LPDM Flexpart [2][3] and the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) Model [4]. In both models forward tracking and inverse (or receptor-based) modes are possible.

Certain distinct pollution episodes have been selected from the database EXPER/PF and also from UK monitoring stations and their likely trajectory predicted using prevailing weather data at the time of discharge. Global meteorological data were downloaded from the ECMWF MARS archive. Preliminary inverse tracking studies indicated that many of the most districted exceedences observed, originated outside the monitoring area. For example, heightened particulate concentrations are shown to originate from sand storms in the Sahara, continental forest fires or even volcanic activity in Iceland and the Caribbean.

One of the principal aims of this project is to make best use of the available receptor data sources in order to evaluate and improve available forecasting models. In the process we aim to investigate the most suitable methodology for accuracy ease of use and computational speed.

This paper continues with a brief description of the methodology employed in this project, in particular the numerical models in the code of Flexpart (Version 6.1) which was kindly provided to us in source form. This is followed by a description of the input data used and examples of typical pollution episodes.

2. Methodology

Aerosol dispersion models generally use meteorological fields and emission inventories as input, and calculate time-dependent concentration fields as output, using suitable models for the relevant processes such as advective or convective transport, mixing by diffusion, and mass transfer through dry or wet deposition, or chemical reaction.

The mathematical models discussed in this paper use input data (meteorological, emissions or receptor data, land use etc) to predict pollutant concentration, or alternatively to identify pollution sources.

Backward simulations can be performed by applying the same methodology and numerical model, but with the particle trajectories being integrated backwards in time, so using a negative time step.

2.1 Particle Dispersion Model "Flexpart 6.1"

Flexpart is a Lagrangian Particle Dispersion (LPD) model, developed by Andreas Stohl (University of Munich). Since it is an open source code (Fortran 77) project it can build a good base for the needs of our simulations.

Flexpart has been validated with measurement data from large-scale tracer experiments, during case-studies of stratospheric intrusions and has performed well in comparison with other similar models [2][3]. In this paper Flexpart will be evaluated with other comparable models such as the HYSPLIT model.

2.2 The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) V.4.7 Model.

HYSPLIT is the result of cooperation between the National Oceanic and Atmospheric Administration (NOAA) and Australia's Bureau of Meteorology. It is a widely used LPD model and it is currently used operationally for emergency response at NOAA. The dispersion of a pollutant within HYSPLIT is calculated by assuming either a Gaussian or a top-hat distribution within a puff or from a fixed particle position. It is coded in Fortran-90 and incorporates its own Graphical User Interface and several tools to analyse and post-process the output of air concentration, trajectory, or precipitation simulations.

2.3 Meteorological Input data

Both models are driven in this project by meteorological fields from the Mars archive of the European Centre for Medium-Range Weather Forecasts (ECMWF), encoded under the WMO FM-92 GRIdded Binary (GRIB) format. The data sets within ECMWF are based on quantities analysed or computed within the re-analysis data assimilation schemes or from forecasts based on these analyses. An authorised user account is granted for this project, which allows for access to the ECMWF computing facilities. The data is retrieved and pre-processed using the ECMWF computing facilities and usually have a horizontal resolution of 0.5 degrees, 60 vertical levels and temporal resolution of 3 hours.

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The UK Air Quality Information Archive

The Air Quality Data and Statistics Database offers the retrieval of statistics related to Air Quality in the UK, from the present back to 1960, from the monitoring networks operated on behalf of the Department for Environment Food and Rural Affairs, UK (DEFRA) and the devolved administrations.

The Kent Air Quality Monitoring Network

are released in prescribed source regions, and transported with the mean and a stochastic turbulent velocity field. Particles can then be tracked in forward and backward mode.

The output of the simulations shows that there are notable differences between the two formulations, although both models use the same meteorological data and source input. This means that the identification of the primary emissions during air pollution episodes may be rather uncertain, highlighting the need to make maximum use of the available air quality data.

Despite the differences, which are under investigation, the simulations give a useful indication of the location of possible sources of the pollution even over long distances and considerable time span.

The results of the two Lagrangian particle models will be evaluated and compared with available monitoring data and satellite images.

To improve the accuracy of the simulations, a higher grid resolution will be adopted in future model runs. In addition, the number of the notional particles will be increased and the models will be driven by weather data of higher resolution also containing additional information such as anthropogenic heat production and diurnal cycle of surface stress over mountainous areas.

One of the difficulties in meso-scale atmospheric modelling in the UK is the limited availability of ambient atmospheric monitoring data for many pollutants of concern. For example, background concentrations of particulate matter are only monitored by few stations (Harwell, Rochester) in the rural automated network of UK.

Acknowledgements

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Appendix 3: Copy of the Presented Paper on the 10th Int. Conf. on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes

Plainiotis S., Pericleous K.A., Fisher B.E.A., and Shier L., 2005: Application of Lagrangian Particle Dispersion models to air quality assessment in the Trans-Manche region of Nord-Pas-de-Calais (France) and Kent (Great Britain). Proceedings of the 10^{th} Int. Conf. on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes, Sissi, Crete, October 17-20, 2005, pp. 398-403

APPLICATION OF LAGRANGIAN PARTICLE DISPERSION MODELS TO AIR QUALITY ASSESSMENT IN THE TRANS-MANCHE REGION OF NORD-PAS-DE-CALAIS (FRANCE) AND KENT (GREAT BRITAIN).

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INTRODUCTION

In the framework of the cross-border EU Interreg IIIA activity, the joint Anglo-French project, ATTMA, has been commissioned to study Aerosol Transport in the atmosphere of the Cross-Channel, or "Trans-Manche" region of Nord-Pas-de-Calais (France) and Kent (Great Britain). The air quality of the region is dominated by the industrial area of Dunkerque, in addition to transportation sources linked to cross-channel traffic in Kent and Calais. The project aims to determine, through modelling pollutant dispersion patterns and identify sources responsible for episodes detected, using ground monitoring data from UK and French networks and with the assistance of satellite images.

Lagrangian Particle Dispersion (LPD) models compute trajectories of a large number of notional particles and can be used to numerically simulate the dispersion of a passive tracer in the planetary boundary layer. The project uses two widely used particle dispersion models: the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model and the model FLEXPART. Both models possess the forward tracking and inverse (or receptor-based) modes. Both models are driven by meteorological datasets output from the MM5 mesoscale model. Datasets from the ECMWF MARS archive are used to initiate the MM5 model. This paper investigates the sensitivity of these models to input data resolution and in particular the impact of mesoscale topographic effects. Previous work by the authors [Plainiotis, 2005a 2005b] concentrated on the identification of far-field sources driven by synoptic conditions.

DATA AND METHODS

A number of requirements must be met for credible air pollution modelling. The mathematical models discussed in this paper use input data (meteorological, emissions etc.) to predict pollutant concentration, or to identify pollution sources.

Particle Dispersion Model "FLEXPART"

FLEXPART is a Lagrangian Particle Dispersion (LPD) model, developed by Andreas Stohl (1998) and is widely used to calculate the transport and dispersion of non-reactive pollutants in the atmosphere [Stohl,1998]. It is an open source code (Fortran 77) project; hence it can provide a good base for the project. FLEXPART has been validated with measurement data from large-scale tracer experiments, during case-studies of stratospheric intrusions and has performed well in comparison with other similar models [Stohl, 1998]. FLEXPART is driven by synoptic data from ECMWF.

The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) V.4.7 Model.

HYSPLIT is an alternative LPD model used in comparisons. Dispersion assumes either a Gaussian or a top-hat distribution within a puff or from a fixed particle position. HYSPLIT is coded in Fortran-90 and incorporates its own Graphical User Interface and several tools to analyse and post-process the output of air concentration, trajectory, or precipitation simulations. The main difference with FLEXPART is in its characterisation of dispersion coefficients. FLEXPART has enhanced physics with vertical diffusion and convection mixing.

Meteorological Input data

Aerosol dispersion models generally use synoptic fields and emission inventories as input, and calculate time-dependent concentration fields as output, using suitable models for physical processes such as advective or convective transport, mixing by diffusion, and mass transfer through dry or wet deposition, or chemical reaction. Several sources used in this project are given below.

The European Centre for Medium-Range Weather Forecasts (ECMWF)

In this project both LPD models can be driven by meteorological data from the Mars archive of the European Centre for Medium-Range Weather Forecasts (ECMWF), encoded under the WMO FM-92 GRIdded Binary (GRIB) format. Scale resolution, 0.36 degrees, 90 vertical levels and 3 hours temporal (as of 19-08-05).

Meteorological fields of higher resolution by the Fifth-Generation NCAR / Penn State Mesoscale Model (MM5), Version 3

The Version 3 of the PSU/NCAR Mesoscale Model, nonhydrostatic, terrain-following and sigma-coordinate model, was designed to simulate or predict mesoscale and regional-scale atmospheric circulation. The model, known as MM5 V.3 is in open-source and contains contributions from users at several universities and government laboratories. It provides a good base for our simulations because of multiple-nesting capability and can output weather data of higher resolution. It is supported by several auxiliary programs, which are referred to collectively as the MM5 modelling system. Input data are required:

Terrain data. MM5 model requires various terrain input data, such as elevation, vegetation/land-use, land-water mask and vegetation. High resolution terrain data of 0.3 degrees (0.925 km) is adopted for this project, available from the USGS EROS Data Centre's anonymous ftp site edcftp.cr.usgs.gov under directory: /pub/data/gtopo30/global.
Analysis Data. Meteorological data from ECMWF, encoded under the GRIB format can be used to provide the initial weather fields for the MM5 V.3 model runs.

The data, encoded under the GRIB format are then re-gridded and interpolated to create an input for MM5. The GRIB data are re-gridded and interpolated to the horizontal grid and map projection as defined by the MM5.

MM5 V.3 model output

FLEXPART in its official version is driven by synoptic ECMWF data. However, modified versions of FLEXPART are available for handling the mesoscale MM5 model, based on older versions of FLEXPART (3.1) [G. Wotawa ,2000].

The MM5V3 output is first converted to the format of MM5V2, using a utility provided by NCAR. The resulting MM5V2 data is converted to ECMWF GRIB format, using the preprocessing tool provided by G. Wotawa. The resulting data use the MM5's Lambert Conformal projection instead of the ECMWF's geographical latitude-longitude system and can provide a direct input for the MM5 version of FLEXPART. In HYSPLIT, the ECMWF GRIB datasets are converted to the model's ARL native format.

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Appendix 4: Technical Notes

1. Installation of the GRIBEX software for decoding the GRIdded Binary (GRIB) encoded data

The GRIBEX and PBIO libraries have been developed by ECMWF for encoding and decoding GRIB code messages and are available for free at the official ECMWF website.

The current version of ECMWF's GRIBEX (220) can be compiled on the following Operating Systems:

CRAY, FUJITSU, VPP5000, DEC ALPHA, DECMIPS, HPPA, i686, IBM, LINUX, RS6000, SGIMIPS, SUN4 .

In order to install the GRIBEX and PBIO libraries, the compressed GRIBEX tar file was first downloaded from the ECMWF ftp directory and extracted to our local Linux machine.

Then, in the GRIBEX source code directory, the compiler was executed using the command:

make ARCH=linux_g77

also indicating the configuration/source files that correspond to the current operating system and the Fortran processor. This compiles a FORTRAN library called 'libemos.a', containing all the subroutines and functions for manipulating GRIB data that will later be linked to the FLEXPART code.

2. Installation of FLEXPART

The source code of FLEXPART is freely available for scientific use at the web site of Andreas Stohl:

http://www.forst.uni-muenchen.de/EXT/LST/METEO/stohl/FLEXPART.html

Once the source is downloaded at the local workstation, it has to be extracted (untared) conFigured and compiled.

If FLEXPART is linked to a recent version of the GRIBEX libraries (e.g. 220), all calls to the swap32 subroutine need to be commentded out, as this function is now completed automatically by the decoder. Furthermore, to enable switching from forward to backward runs some parts of FLEXPART's code have to be changed, to improve the efficiency of memory use. For the simulations of our project, two different versions of FLEXPART were compiled, one for forward and one for backward runs.

FLEXPART, by default, requires the availability of considerable amounts of memory and computational computing power to execute the model. However it can be optimised for use on almost any (relatively recent) UNIX workstation.This can be achieved by changing the default limits for the input data, such as the wind fields, number of particles, or grid dimensions.

The following parameters of the FORTRAN77 file `includepar` were adjusted for our installation:

1. maxpart (number of particles) ,

2. maxpoint (number of release locations/receptor points in backward mode)

3. maxspec (number of chemical species per release)in the Fortran file maxwf (number of wind fields to be used for simulation)
4. maxtable (number of different chemical species)
5. numclass (number of landuse classes available to FLEXPART)
6. ni (Number of diameter classes of particles)

For example in the forward mode the following configuration was made:

parameter(maxpart=2000000,maxpoint=200,maxspec=1)

parameter(maxwf=50000,maxtable=1000,numclass=9,ni=11)

The backward mode usually requires higher amounts of RAM, so narrower limits should be chosen;

parameter(maxpart=200000,maxpoint=12,maxspec=1)

parameter(maxwf=50000,maxtable=1000,numclass=9,ni=11)

After the adjustments are made, the source code needs to be linked and compiled.

The makefile is changed to reflect the current Fortran 77 compiler's options and linked to the GRIBEX decoding libraries (file libemos.a).

For our g77 compiler the configuration was changed to:

FFLAGS = -O2 -C -P -B108 -B101 -U -N86

LDFLAGS = -O2 -C -P -X -static -B108 -B101 -U -N86 -lg2c /path_to_gribex/libemos.a

3. Configuration of FLEXPART

Characteristics of the model run are specified within the files in the directory "Options", of the default FLEXPART installation directory.

The main files are:

COMMAND, RECEPTORS, RELEASES, SPECIES, OUTGRID

The file COMMAND specifies:

- the simulation mode, forward or backward
- the start and the end time of the simulation,
- the frequency Tc,
- the averaging time DTc and
- the sampling intervals DTs of the output fields,
- the time constant for particle splitting Dts,
- the synchronisation interval of the model ,
- the factor by which the time steps must be below the Lagrangian time scale ctl,
- the refinement factor for the Langevin equation for the vertical turbulent wind component ifine.

The file RECEPTORS specifies the locations of the receptor points for which more detailed air concentrations is calculated.

File RELEASES determines the release specifications (or the pollution measurement data in the backward mode).

4. Preparation of ECMWF GRIdded Binary (GRIB) data, prior to July 2004

This paragraph provides a detailed technical description and a step-by-step setup guide for original routines developed Gerhard Wotawa and Paul James for the preparation and retrieval of ECMWF GRIdded Binary data. The routines are freely available at Paul James' web site: http://www.forst.tu-muenchen.de/EXT/LST/METEO/stohl/FLEXTRA/ecmwf_extr.html

These routines are not longer compatible with the new ECgate system of ECMWF (after July 2004) and are no longer supported or maintained by G. Wotawa. The new version of retrieval routines by G. Wotawa, *et al* in July 2004 (section 3.4.2) were developed entirely from scratch, hence they have different methodology and require a different setup procedure.

However, this paragraph can be particularly useful for FLEXPART users who want to pre-process data using our own data retrieval routines available at the website of our research (www.gre.ac.uk/~ps60) which are mostly based on the original routines of Wotawa but have been ported to the new ECgate server. As can be seen in paragraph 3.4.3, these maintained original routines offer various advantages over the second version of G. Wotawa routines. Also, this paragraph describes the data preparation procedure followed in many of the case studies presented.

The routines perform the following tasks:

a) Retrieval of the accumulated flux data from the MARS archive (e.g. precipitation, radiation, sensible heat flux),

b) De-accumulation of flux data, storage of results in the ecfs file-system,

c) Retrieval of other meteorological fields

d) Calculation of the vertical velocity component d(eta)/dt

The following paragraphs give a step-by-step guide to the above procedure.

After the routines are uploaded to the home user directory of the remote file system of ECMWF, they have to be extracted and conFigured to reach the correct paths to the account's directories. Also the type of required data should be conFigured, such as the region's co-ordinates and the beginning/ending date of the simulation. The routines for tasks 1 and 2 can be found in any of the directories:

fluxjobs_4dvar, fluxjobs_era (for) , fluxjobs_gen.

In each case the user has to choose which of the above directories corresponds to the required data type and period, for example the routines at fluxjobs_era prepare data of the ERA-15 type. For data from year 2000 and beyond, the directory fluxjobs_4dvar can be used and hence it was the preferred one for the project's simulations.

A) Retrieval of the accumulated flux data

First new directories should be created in the scratch filesystem to accommodate the flux data. This can be done after the user has connected to the remote ECgate system by the UNIX command:

mkdir $SCRATCH/flux

The FORTRAN include file "parameter" has to be modified during each data retrieval, according to the specifications of the required grid, such as:

nx, ny: number of grid points

xlon0, ylat0: longitude and latitude of lower left grid point

dx, dy: grid distance

Also the following scripts have to be conFigured: surfjob.new for the retrieval of the accumulated flux data from the MARS archive and deacc.flux for the deaccumulation of the fluxes. Then all the routines have to be compiled remotely with the FORTRAN compiler using the command:

f77 *.f $EMOSLIB

Then the binary output has to be renamed to FLUXACC , so that it can be run later by the scripts:

mv a.out FLUXACC

After all configurations are completed, the script surfjob.new is put in batch queue by executing the command:

qsub -q ecgate1.normal surfjob.new

The following flux data, needed by FLEXPART, can be retrieved on a user-selectable geographical grid (lamda-phi) (Wotawa , *et al* 2003):

Abreviation	**Type of Data**	**Unit**
LSP	Large Scale Precipitation	mm/hr
CP	Convective Precipitation	mm/hr
SSHF	Surface sensible. heat flux	W/m^2
EWSS	East-west surf. stress	N/m^2
NSSS	North-South Surf. Stress	N/m^2
SSR	Surf. Solar Radiation	W/m^2

B) De-accumulation of FLUX data

When the accumulated flux data is retrieved from the MARS archive, the script deacc.flux is executed to perform the de-accumulation of fluxes and copy the resulting files to the scratch filesystem.

The de-accumulation of precipitation (LSP, CP) data, is based on the following methodology:

- Accumulated values for 3/6/9/... UTC are just divided by the number of hours, namely 3.
- Accordingly, precipitation at X UTC is the mean precipitation per hour from X-3 to X UTC

For the de-accumulation of the other flux quantities the following strategy is used:

Accumulated values are divided by the number of hours and also interpolated to 3/6/9... UTC (for a three-hourly first guess with evaluation time X UTC, a flux value has been accumulated from X-3 UTC to X UTC and is therefore valid for X-1.5 UTC)

Accordingly, radiation (X UTC) is the radiation valid for X UTC (three-hourly average)

C) Retrieval of other meteorological fields and calculation of the vertical velocity component d(eta)/dt

These tasks are performed by the routines found at any of the directories: job_era, job_pre99, job_y2k, again according to the time period of the required GRIB data. Here the CONTROL file has to be edited, providing information like beginning date, ending date, time interval, number of levels to extract, scratch directory for (optional) flux data input, ecfile directory for output.

Afterwards, the FORTRAN sources are compiled **(f77 *.f $EMOSLIB).**

The execution of the resulting binary creates a batch script, which sends to the batch queue of the Mars archive and executes the routine CONVERT_PRE, which:

- Reads all ECMWF model data.
- Makes the wind fields 3D mass consistent, by calculating the vertical velocity (gridded values) component in the ECMWF eta vertical co-ordinate system (d(eta)/dt) applying the equation of continuity.
- Writes out all meteorological data including d(eta)/dt
- Saves the resulting output files (ECMWF GRIB format) in the user-specified directory of the EC File System.

5. Numerical reciepies in FLEXPART

i) Dispersion calculation

FLEXPART, like most Lagrangian particle dispersion models assumes the Turbulent motions v_t for wind components i to evolve as a Markov process (Wilson and Sawford, 1996) according to the Langevin equation (Thomson, 1987):

$$dv_{t_i} = \alpha_i(x, v_t, t)dt + b_{ij}(x, v_t, t)dW_j \qquad (1),$$

where the drift term α and the diffusion term b are functions of the position x, the turbulent velocity and time. dW_j are incremental components of a Wiener process with mean zero and variance *dt*, which are uncorrelated in time (Legg and Raupach, 1982, adapted from Stohl, *et al* 2005).

Uliasz (1994) has indicated that cross-correlations between the different wind components have little effect for long-range dispersion, hence they are not taken into account. The particle position increments are calculated from:

$$dx_i = u_i dt \quad (2),$$

Gaussian turbulence is always assumed in FLEXPART. Although this assumption is valid under stable and neutral conditions, it is violated under convective conditions when turbulence is distorted and larger areas are occupied by downdrafts than by updrafts. However, for transport distances where particles are rather well mixed throughout the atmospheric boundary layer (ABL), the error is minor (Stohl *et al*, 2005). FLEXPART applies a modification to the ABL height (for the equation see below) according to Vogelezang and Holtslag (1996) using the critical Richardson number concept. The bulk-Richardson number is the ratio of convective available potential energy (CAPE) to the magnitude of low-level shear; it measures the intensity of turbulence.

With the above assumptions, the Langevin equation (1) for the vertical wind component ω can be written as:

$$d\omega = -\omega \frac{dt}{\tau L_\omega} + \frac{\partial \sigma_\omega^{\ 2}}{\partial z} dt + \frac{\sigma_\omega^{\ 2}}{\rho} \frac{\partial \rho}{\partial z} dt + \left(\frac{2}{\tau L_\omega} \right)^{1/2} \sigma_\omega \, dW \qquad (3),$$

where ω and σ_ω are the turbulent vertical wind component and its standard deviation, τL_ω is the Lagrangian timescale for the vertical velocity autocorrelation and ρ is density.

The term $\frac{\partial \sigma_\omega^{\ 2}}{\partial z} dt$ is the drift correction (McNider *et al.*, 1988) and the term $\frac{\sigma_\omega^{\ 2}}{\rho} \frac{\partial \rho}{\partial z} dt$ is the density correction (Stohl and Thomson, 1999).

The expression of the Langevin equation of (3) is identical to the one proposed by Legg and Raupach (1982), except for the term from Stohl and Thomson (1999) which accounts for the decrease of air density with height.

If the Langevin equation is expressed in terms of $\frac{w}{\sigma_\omega}$ instead of wthen it becomes (Wilson *et al.*, 1983):

$$d\left(\frac{w}{\sigma_\omega} \right) = -\frac{\omega}{\sigma_\omega} \frac{dt}{\tau L_\omega} + \frac{\partial \sigma_\omega}{\partial z} dt + \frac{\sigma_\omega}{\rho} \frac{\partial \rho}{\partial z} dt + \left(\frac{2}{\tau L_\omega} \right)^{1/2} dW \qquad (4),$$

Thomson (1987) has shown that this expression of the Langevin equation is seen to fulfil the well-mixed criterion of Rodean (1996) which proposes that "if a species of passive marked particles is initially mixed uniformly in position and velocity space in a turbulent flow, it will stay that way".

Although the method proposed by Legg and Raupach (1982) (3) violates this criterion in strongly inhomogeneous turbulence, their formulation was found to be practical, as numerical experiments have shown (Stohl, 2005) that it is more robust against an increase in the integration time step. Therefore, (3) is used in FLEXPART where longer time steps are needed; otherwise, (4) is used instead (Stohl *et al* , 2005).

ii) Removal processes

FLEXPART takes into account radioactive (or other) decay, wet deposition, and dry deposition by reducing a particle's mass.

Radioactive decay is accounted for by reducing the particle mass according to

$$M(t + \Delta t) = M(t) \exp(-\Delta t/\beta) \qquad (6),$$

where m is particle mass, and β the time constant=$T_{1/2}$/ ln(1/2) is determined from the half life $T_{1/2}$, specified in file SPECIES. Deposited pollutant mass decays at the same rate. **Dry deposition** can be defined as the process of the removal of particles and gases from the atmosphere due to turbulent transfer or gravity separation and subsequent uptake or deposition at the surface.

The dry deposition velocity of a gas is calculated with the resistance method (Wesely and Hicks, 1977) according to:

$$|\upsilon_d(z)| = [r_a(z) + r_b + r_c]-1 \qquad (7),$$

where r_a is the aerodynamic resistance between z and the surface, r_b is the quasilaminar sublayer resistance, and r_c is the bulk surface resistance. The deposition of particulates is calculated according to:

$$\upsilon_d(z) = [r_a(z) + r_b + r_a(z)\, r_b\, u_g]^{-1} + u_g \qquad (8),$$

where u_g is the gravitational settling velocity calculated from (Slinn, 1982):

$$u_g = \frac{g\rho_p d_p{}^2 C_{cun}}{18\mu} \qquad (9),$$

where ρ_p and d_p are the particle density and diameter, μ the dynamic viscosity of air (0.000018 kg/ms) and C_{cun} the Cunningham slip-flow correction.

In addition, **wet deposition** that is removal due to precipitation can be calculated in FLEXPART using scavenging coefficients. In principle, in-cloud and below-cloud scavenging must be separated [(Asman, 1995), adapted from (Stohl *et al,* 2005)]. However, as data on cloud base height and depth are not available, in-cloud and belowcloud scavenging are treated jointly in FLEXPART and wet deposition takes the form of an exponential decay process:

$$m(t + \Delta t) = m(t) \exp(-\Lambda\Delta t) \qquad (10),$$

where m and Λ are the particle mass and the scavenging coefficient, respectively. The scavenging coefficient 3 increases with precipitation rate according to

$$\Lambda = AI^B \qquad (11),$$

Information and characteristics about the wet and dry deposition behaviour of gases and particles should be provided by the user in the configuration file SPECIES (see appendix). However a constant velocity of dry deposition can be specified to be used by FLEXPART if detailed information is not available.

If dry deposition of particles is set to be calculated, data on land uses is required. FLEXPART includes land use data for the European Region, taken from a database developed by van de Velde *et al.* (1994) which provides the area fractions of eight land use classes on a grid with 10' resolution.

iii) Inverse mode

Inverse simulations can be performed by applying the same methodology and numerical model, but with the particle trajectories integrated backwards in time, hence using a negative time step. The theory of modelling dispersion backward in time with Lagrangian particle models was developed by Flesch *et al.* (1995) and Seibert and Frank (2004). Many experiments and examples have shown the ability of FLEXPART (running in inverse mode) to determine linear-source receptor relationships for the transport of atmospheric trace substances (Seibert *et al*, 2001), (Stohl *et al*, 2003). The derivation includes the action of sources and of any first-order processes (transformation with prescribed rates, dry and wet deposition, radioactive decay etc).

iv) Calculation of concentrations

The ensemble of individual Lagrangian particles in a grid volume needs to be converted to a mass of volume concentration. FLEXPART offers the choice of output either in the form of concentrations, volume mixing ratios or both. It calculates concentration in a grid cell by sampling the tracer mass fractions of all particles in the grid cell and dividing by the grid cell volume:

$$C_{T_S} = \frac{1}{V}\sum_{i=1}^{N}(m_i fi) \qquad (12),$$

Where V is the grid cell volume, m_i is the particle mass, N the total number of particles, and f_i the fraction of the mass of particle i attributed to the respective grid cell. This mass fraction is calculated by a uniform kernel with bandwidths (Δx, Δy), where Δx and Δy are the grid distances on the longitude-latitude output grid (Stohl, 2005).

v) Uncertainties

FLEXPART estimates the uncertainty of the output by carrying many (default value of 10) different classes of particles in the model simulation and calculating the concentration individually for each class. Then, at every grid point the deviation from every concentration is calculated. Relative uncertainty derives from the standard deviation divided by the mean concentration. In contrast HYSPLIT does not provide a direct way to determine the uncertainty of the output.

vi) Model output

FLEXPART output is produced in gridded unformatted binary format. Tracer concentrations and/or mixing ratios (for forward runs), or emission sensitivity response functions (for backward runs) are calculated on a 3-D longitude-latitude grid, defined in the configuration file OUTGRID (see appendix 4). The output domain and resolution can differ from the grid on which meteorological input data are given. Two-dimensional wet and dry deposition fields are outputted over the same spatial domain, and tracer mass fluxes can also be determined on the 3-D grid. For certain locations, specified in the configuration file RECEPTORS (see appendix 4) and independently from a grid, time series of concentrations can also be outputted.

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