Reviewing Issues Associated with Modelling Atmospheric Dispersion in Changing Meteorological Conditions

A report prepared for ADMLC by

Christelle Escoffier, Françoise R. Robe, Alfred M. Klausmann and Joseph S. Scire

TRC Environment Corporation

ABSTRACT

Changing meteorological conditions along the path of a pollutant between source and receptor are non-steady-state conditions by definition. Temporal and spatial changes in meteorological conditions cover a broad spectrum from large scale (large cyclonic or anticyclonic system) down to local scale (such as micrometeorological variations). Steady-state models, such as Gaussian plume models, are widely used for atmospheric dispersion simulations for different applications and in many cases are adequate for the purpose. However, depending on the distance from the source to the point of interest for a specific application, the rapidity of the meteorological changes and the type of application considered, the use of a steady-state model could be questioned.

The review identified the typical changing meteorological conditions which could trigger non-steady-state situations and impact on the pollutant concentration (accumulation, recirculation or deposition) and/or the pollutant path (curved trajectory). The assumptions of steady-state models are discussed to determine situations when the simulation should use a non-steady-state model instead. Most of the discussion being qualitative, datasets from experiments which could be used for testing the sensitivity and the discrepancy between steady-state and non-steady-state models in critical non-steady-state situations are identified. A number of tests are proposed to quantify the discrepancies in those specific situations.

This study was funded by the UK Atmospheric Dispersion Modelling Liaison Committee.

EXECUTIVE SUMMARY

This review, commissioned by ADMLC, investigates the modelling of atmospheric dispersion in variable weather conditions and their impact on various applications. The focus of the study was to look at the effects of temporal changes in meteorological conditions on the pollutant plume when travelling between source and receptor. Steady-state models, such as Gaussian plume models, are widely used for atmospheric dispersion simulations for different applications and in many cases are adequate for the purpose. However, results of some non-steady-state situations simulated by steady-state models should be treated with caution and the use of a steady-state model may be questioned.

The review identified the typical changing meteorological conditions which could trigger non-steady-state situations and impact adversely on the pollutant concentration or deposition fluxes (accumulation, recirculation or deposition) and/or the pollutant path (curved trajectory). The main changes in meteorological conditions investigated are passage of fronts, low wind speed conditions, thermally induced circulations and temperature inversions but it is worth noting that any combinations of these conditions can occur regularly. Some variations in meteorological conditions are defined by the local physical characteristics of an area, such as land/sea breeze circulation or mountain/valley circulation, and can occur only at specific locations but others, such as the passage of fronts, low wind speed or temperature inversion phenomena, can occur in any location.

Dispersion modelling covers a large range of applications: accidental release, emergency response, risk assessment, regulatory impact assessment, operational real-time and forecasting. The end point of interest could be more sensitive to the pollutant concentration, the path followed by the pollutant plume or a combination of both. The timescale can vary from sub-hourly or hourly to seasonal or an annual averaging time. These requirements were discussed for each application and evaluated against timescales of changes in meteorological conditions. The possible non-steady-state situations encountered by each application are discussed to indicate whether steady-state models are adequate and when their use should be questioned. Indeed, depending on the distance from the source to the location of interest for a specific application, the rapidity of the meteorological changes and the type of application considered, the use of a steady-state model may not be appropriate.

The diversity of models is discussed from simple and advanced Gaussian plume models to Lagrangian and Eulerian non-steady-state models. The assumptions of steady-state plume models are compared to the assumptions of non-steady-state models to identify the non-steady-state situations when steady-state models are not adequate. The three main characteristic differences between Gaussian plume models and Lagrangian non-steady-state models are (i) travel to infinity versus fixed finite travel distance, (ii) not remembering versus remembering the previous several time steps footprint and (iii) single point wind

data versus three dimensional wind fields. All could have an effect on the location and concentration of the highest peaks. Any applications which are sensitive to the exact location and/or amount of pollutant predicted display large discrepancies when using a simple Gaussian plume model versus a non-steady-state Lagrangian puff or particle model. The shorter the time average impact the user is interested in, the stronger the discrepancies are.

Steady-state models are usually appropriate for modelling pollution impact at mesoscale distances from a continuous-release source provided the land characteristics are spatially constant between the source, the receptors and the meteorological stations involved in the modelling, and the flow remains non complex. However, when the meteorological conditions are changing rapidly at a given location within the domain or when they are changing spatially within the domain, the accuracy of steady-state dispersion modelling for predicting the changes in dispersion when the outcome is on a short-term timescale could be questioned.

The distance from source where the conditions become non-steady-state is an important parameter to identify in air dispersion simulation using steady-state models. It is dependent on the source characteristics but also land surface conditions and meteorology. A steady-state index, described in this review, which is computed to quantify the difference in meteorological variability at source and at receptor locations could help to determine how far from the source steady-state conditions remain valid.

Availability of experimental datasets which could be used for testing the sensitivity and the discrepancy between steady-state and non-steady-state models in critical non-steady-state situations are discussed. A vast variety of experimental datasets is available but not all are suitable. The most adequate for testing the sensitivity to changes in meteorological conditions are the long-range tracer experiments. However, they involve distances from source that are not compatible with steady-state model application, especially where short-term averages are of interest. Nevertheless, a few experimental datasets have been identified as useful for sensitivity testing of steady-state models versus non-steady-state models in a number of changing meteorological conditions such as land/sea breeze, breaking up of morning temperature inversion, passage of fronts and low wind speed conditions. A list of sensitivity tests using these datasets are described and proposed to be developed in future work to help quantify the discrepancies between models and provide additional guidance regarding atmospheric dispersion modelling in changing meteorological conditions in those specific situations.

CONTENTS

1 INTRODUCTION

This review, commissioned by ADMLC, investigates the modelling of atmospheric dispersion in variable weather conditions and their impact on various applications. A number of atmospheric dispersion models used for dispersion modelling applications are steady-state models, which do not allow any temporal changes in meteorology on the pollutant path between the source and the receptors of interest. The characteristics of most of these models prevent them from simulating a curved trajectory or from applying any changes after the first time step of the pollutant release. Changing meteorological conditions affecting pollutants transported between a source and the receptors of interest are non-steady-state conditions by definition. Depending on the application and its requirement, the use of a steady-state model might still be adequate, indeed in many situations, the distance and time scale for the pollutant to travel from the source to the receptors are such that the conditions can be considered quasi-steady-state. However, this review aims to highlight under which meteorological circumstances the impact on a range of dispersion applications would be significantly different if simulated with a steady-state or a non-steady-state dispersion model.

Here is a list of key points we aim to address in this review:

a What are the types of temporally changing meteorological conditions and their causes
b What are the temporal and spatial scales of the variable weather conditions
c How the changing meteorological conditions can affect atmospheric dispersion
d What are the requirements of dispersion modelling for various applications
e How different is the impact of dispersion applications if modelled with a steady-state or non-steady-state model in variable weather conditions
f For which time scales and situations the simulation of changing meteorological conditions with steady-state models is appropriate
g Are there alternative models and suitable datasets for evaluating models in changing weather conditions
h Suggestion of a number of tests designed to quantify the qualitative discrepancies of modelling impacts

Initially, changing meteorological conditions and their causes are identified and listed using examples taken from the literature. The emphasis is put on examples showing how the changes in meteorological conditions can have adverse effects on pollutant concentrations. Secondly, a variety of dispersion modelling applications is discussed. Particular attention is directed to the requirements of those applications and their outcomes. Historical events and some existing datasets are examined where relevant. In the third part, we aim to identify what steady-state and non-steady-state means in terms of atmospheric dispersion modelling. Timescales of the meteorological events and dispersion applications are cross-referenced and a discussion of which situations can be considered as steady-state or non-steady-state depending on the type of application is developed. Temporal changes are the focus of the report, but spatial changes are difficult to ignore since the path between the source and the receptor is the subject of interest. Spatial changes are also discussed together with calm wind conditions and source characteristics.

The choice of atmospheric dispersion models is vast. Discussing the availability of suitable steady-state and non-steady-state models and field experiments datasets to assess the consequences of using a steady-state model for a non-steady-state application is developed in the last section of our review. It is underlined that the meteorological data input into the models whether as a one-dimensional field or as a three-dimensional field can make a difference for the modelling of changing meteorological conditions on the pollutant path between the release and the receptors. A series of sensitivity tests is proposed to be developed in future work to quantify the potential discrepancies between steady-state and non-steady-state simulation in a selected number of changing meteorological conditions leading to adverse effects on pollutant concentrations in mostly near-field applications on short-term timescales.

However, an exhaustive review is beyond the scope of this work since the number of combinations of changing meteorological conditions with type of atmospheric dispersion applications, and type of atmospheric dispersion models is unlimited.

2 CHANGING METEOROLOGICAL CONDITIONS

Various types of changing meteorological conditions are discussed in this section, ranging from large scale cyclones to localized temperature inversion-breakup. Emphasis is placed on meteorological processes that lead to temporal changes in meteorological parameters which are important for the outcome of the dispersion of pollutants, such as wind speed and direction, moisture content, atmospheric stability, turbulence and mixed layer depth. The relative scale of meteorological events has been put in perspective by a paper by Orlanski in 1975. He proposed a subdivision of scales that covers the entire spectrum of atmospheric processes. His review shows how temporal scales and spatial scales are linked as it is displayed in Figure 1 extracted from Orlanski (1975). All meteorological events described below can be referred to this table for an interpretation of their temporal and spatial scale of influence.

The changing meteorological conditions, illustrated through a number of examples showing their impact on pollutant concentrations, have been divided into two large sections. The first section deals with weather patterns linked directly to the general atmospheric dynamic. On the other hand, the second section shows how the local influence of land use and topography can bring changes in meteorological conditions as well. The last part of this section underlines the complexity of the system and how weather patterns and local thermal influences can interact to create changing meteorological conditions and lead to high pollution events locally.

2.1 Atmospheric Dynamic

Large scale atmospheric circulation systems such as anticyclones (or Highs) and cyclones (or Lows) can cause temporal changes in wind speed and direction on a large range of timescales from hours to days or more. In anti-cyclonic situations, the changes in meteorological conditions are gradual and may be approximately uniform on a relatively large area (100 km^2 to 200 km^2). From one time step to the next, the spatial scale of the temporal changes would usually cover an entire mesoscale domain and a steady-state model would be able to represent such a change. Highs are usually moving at a lower speed than Lows and can also become stationary. Fronts embedded in Lows present much sharper non-steady-state discontinuities and are often accompanied by unstable weather and heavy precipitation. Highs and Lows events are discussed in the two sections below.

2.1.1 Highs

2.1.1.1 Light Winds Dispersion Conditions

Anticyclones can linger over an area for several days. An example of a stationary anticyclone is depicted on Figure 2 where a High can be seen centred on Norway for a 4-day period, January 9 to January 12, 2010. Stationary anticyclones can lead to prolonged periods of light wind speed conditions. Under these calm conditions the wind direction is either poorly defined or determined by turbulence, resulting in rapid and random wind direction changes on a sub-hourly time scale. These conditions can lead to a gradual build-up of pollutant concentrations due to lack of pollutants being transported outside the region of interest. The travel time between the source and the receptor and the memory of the previous time step are both important factors to consider in these situations. Both are characteristics of non-steady-state dispersion conditions, even though the meteorology itself appears as quite steady on hourly (or even longer) basis.

In such conditions, there is a diurnal change in mixing height, the top of the boundary layer. It is low at night when the ground cools down and increases in the morning to reach a peak height at mid-day by the increase in turbulence due to solar radiation heating of the ground. A number of advanced steady-state models, such as AERMOD (US EPA, 2003) or ADMS (Davies et al., 2007) can usually simulate this type of change in meteorological conditions, since it corresponds to progressive changes.

2.1.1.2 Subsidence Inversion

Anticyclones generate subsidence over large areas. The air is heated by compression creating an elevated temperature inversion known as a subsidence inversion. Subsidence inversions in stationary anticyclone situations are persistent and allow build-up of pollutants over a long period of time.

Scire and Chang (1991) studied such occurrences, using field measurements from the South-Central Coast Cooperative Aerometric Monitoring Program (SCCAMP 1985). High ozone pollution events were shown to be correlated with peak 850-mb temperatures, strong vertical stability and overall limited mixing conditions, especially in the months of September and June (see Figure 3, Table 5 extracted from Scire and Chang, 1991).

Previous studies by Smith (1984) and Moore and Reynolds (1986) also found that high ozone concentrations in Ventura County were associated with a high temperature at 850mb and compounded by weak sea-breeze conditions bringing even colder air under the inversion.

In the study of Particulate Matter (PM) pollution events over Northern Greece (Triantafyllou, 2002), the highest frequency of pollution events in the area occurred during a high-pressure system covering the Balkan area. Weak winds are observed on the surface and local thermal circulations are developed. Such conditions result in very stable conditions and the accumulation of pollutants. This situation of subsidence inversion especially occurs during the cold period of the year, and leads to high local concentrations of PM.

2.1.2 Lows and Fronts

More abrupt changes in meteorological conditions over short periods of time can occur during frontal passages. Fronts are usually associated with Lows, which are typically formed by a warm front followed by a cold front. Cold fronts and warm fronts result in marked changes of wind direction, wind speed, temperature and moisture. Fronts also affect the vertical structure of the lower troposphere and create vertical wind shear. Fronts are associated with changes of cloud cover, precipitation and instability. An example of a passage of a Low is shown on Figure 4, it moved from the South West to the North East area of Europe. It was located north of the Canary Islands on February 27, 2010 at 00UTC. It reached the west coast of France on February 28, 2010 at 00UTC causing large scale flooding and damage due to strong wind and precipitation and then moved across the north part of Germany and Finland during the two following days.

2.1.2.1 Spatial Gradients

Fronts are not exactly sharp discontinuities, but rather fairly narrow transition zones with sharp spatial gradients in meteorological conditions. The transition regions associated with fronts can range from about 50 km up to a couple of hundred kilometres. If a front passes over a dispersion modelling domain, the conditions in the domain are not steady-state: temperature, cloud cover and moisture spatial variability at the front cause horizontal gradients of mixing heights between the sources and receptors, while wind shifts across the fronts define a change in plume trajectories also between the sources and receptors. The meteorological conditions change much faster as the Low centre passes above an observer (within hours) than if the observer is located at 50km or more away from the centre where the conditions can stay uniform for a day or more and over hundreds of kilometres.

2.1.2.2 Propagation

The spatial gradients move as the front makes its way across the domain. Cold fronts can move up to twice as fast and produce sharper changes in weather than warm fronts. Since cold air is denser than warm air, it rapidly replaces the warm air preceding the boundary. If a cold front catches up to a warm front, it creates an occluded front, which may increase storm intensity in the area. As it is shown on Figure 4, the warm front and cold front were completely separated on 28 February, while on 29 February the cold front caught the warm front, creating an occlusion. Front propagation and occlusion instability are non-steady phenomena.

2.1.2.3 Precipitation

A cold front commonly brings a narrow band of precipitation that follows along the leading edge of the cold front. These bands of precipitation are often very strong and can bring severe thunderstorms. In British winter and autumn, cold fronts rarely bring severe thunderstorms, but are known for bringing heavy and widespread rainstorms. In the UK, rapid change of weather occurs at all seasons. The west of the country is usually wetter than the east. Extremes of weather occur in the mountains of Scotland, Wales and northern England. At altitudes exceeding 600m, annual rainfall can exceed 1,500 mm and can reach as much as 5,000 mm in some places.

Warm fronts on the other hand are associated with extensive cloud cover and sometimes stratiform rainfall but typically not deep convective showers.

Convection, whether dry or precipitating, often associated with changing winds, is non-steady-state and affects dispersion very much. Patchy cloud cover creates gradients of solar radiation, affecting turbulence and mixing heights, while rain showers form pockets of wet deposition. In opposition, stratiform cloud cover and stratiform precipitation however might be pretty much steady-state over the area they cover.

2.1.2.4 Examples

A number of examples can be found in the literature showing high pollution events associated with the passage of a front.

Lopez et al. (2002) have shown that the Mexico Basin periodically experiences windblown dust events that cause exceedances of the national ambient air quality standard for PM_{10} in the densely inhabited areas of the Mexico City Metropolitan Area. Those high dust episodes are associated with moderate to high winds occurring in early spring when temperatures are high and humidity is low for this region.

In the study performed by Triantafyllou et al., 2002 over the Kozani area in Northern Greece, about a quarter of the high pollution episodes were associated with high wind speed conditions due to the passage of a cold front. The high winds resulted in dust re-suspension and high concentrations of particulate matter.

The frequent passage of the “Sharav” cyclones over the Tel Aviv area during spring causes natural dust outbreaks with extreme values that result in a much higher PM_{10} annual mean in Tel Aviv than in other larger cities in Asia and Europe (Dayan et al., 2005)

The passage of a front is also often associated with precipitation which affects pollution through wet deposition. Dramatic outcomes such as potent acid rain that burns lawns and tree leaves are possible results of such change in meteorological conditions when the front reaches an area with high sulphur dioxide emissions from industries. In 1978 in Wheeling, West Virginia, rainfall acidity was measured at a PH of 1.5-2, the most acidic rain recorded yet, and 5000 times more acidic than normal rainfall (which has a PH ~5).

Dayan and Lamb (2007) have shown how the magnitude of sulphate deposition varies spatially across a region and temporally by season and from year to year. However inter-annual variations of precipitation are most likely linked to larger-scale variations in atmospheric circulation rather than mesoscale and synoptic scale circulations such as fronts and cyclones (Dayan and Lamb, 2005).

2.1.3 Thunderstorms and Squall Lines

A squall line is most simply defined as any line or narrow band of active thunderstorms. The line can extend hundreds of kilometres in length and last for several hours. Because of their long-lasting and well organized convective nature, squall lines are frequently observed to produce heavy rainfall and severe weather events.

One of the characteristics of convective showers, thunderstorms and squall lines is the gust front or outflow boundary. Outflow boundaries are the result of cold downdrafts that spread out laterally at the Earth's surface. For isolated thunderstorms, outflow boundaries can occur over small spatial and temporal scales (a few kilometres over a couple of hours) while squall line outflow spreads out over larger regions (of the order of 200 kilometres) and lasts up to 24 hours.

Outflow boundaries are typically associated with temperature drops, pressure rises, and wind shifts as the boundary passes an observer. The quick changes in meteorological conditions during the passage of the outflow boundary characterize a non steady-state situation.

Thunderstorms and squalls are of course highly convective weather events which not only affect dispersion by their outflows, but also by their vertical motions, unstable conditions and rainfall, all of which are very much non-steady-state and affect pollutant transport and dispersion.

An example of such outflow has been observed and described by Bowen (1996). Figure 6 (Figure 2 from Bowen, 1996) displays vertical profiles while Figure 5 (Figure 3 from Bowen, 1996) shows time series of meteorological variables such as winds, temperature, dew point, sigma-θ, sigma-Φ, etc..., during the passage of a thunderstorm outflow. The conditions before the outflow episode are unstable with strong solar radiation, light winds and large values of horizontal and vertical turbulence at both 12 metres and 92 metres above ground (Figure 5). When the outflow episode arrives, the sudden change in wind direction is accompanied by a large increase in wind speed at 120 metres (from 3.8 m/s at 12.30 LST to 16.4 m/s at 14.30 LST) as shown on Figure 6 b. At the same time, the turbulence drops significantly. The passage of the outflow lasts just a few hours.

2.2 Thermally Induced Circulations

Other changes in meteorological conditions are associated with differential thermal forcing at the earth surface. Thermally induced circulations arise from thermal gradients generated by spatial heterogeneities of surface characteristics. Differential heating notably occurs between land and water boundaries, sloped surfaces and valleys, urban and rural areas, wet and dry soil, snow covered and snow free areas, cloudy and sunny regions.

The related weather patterns vary throughout the day, are localized and can move spatially within the domain. These situations are non-steady-state. Examples below show how the thermally driven (re)circulations can modulate pollutant concentrations.

These thermally-driven changing meteorological conditions can become locally dominant when large-scale winds are weak, such as in high pressure anti-cyclonic conditions.

2.2.1 Land-Sea Breeze

Differential heating over land and over sea (owing to the higher heat capacity of water) results in pressure gradients and land-sea breeze circulations at coastal locations. During the day, the land heats up faster than the ocean, first creating an offshore flow above the surface (at 100m or so), raising surface pressure offshore, which in turns generates a surface onshore flow. After that, the continuous heating over land keeps the sea breeze coming, first directly onshore (i.e. perpendicular to the coast) but soon after onset, the winds veer to the right with time in the Northern Hemisphere owing to the Coriolis effects (e.g. a northerly sea breeze at the onset turns into an easterly sea breeze by dusk). The larger the temperature contrast and pressure gradient, the stronger the associated sea breeze circulation: sea breezes (onshore) are at their maximum during hot spring days and early summer days when the seas have not warmed up yet. Conversely, in late autumn and early winter, the seas have not cooled too much yet and strong land breeze (offshore winds) can develop, with winds flowing from the cold land towards the warmer seas, especially at night when the thermal contrast is the strongest. During low synoptic wind periods, a land-sea breeze recirculation pattern can develop when the ocean temperature is lower than daytime land temperatures and higher than night-time land temperatures. Daytime onshore winds are then replaced by (generally) weaker offshore winds at night. Pollutants are then flushed offshore at night and trapped in the stable marine boundary layer, and brought back in the morning by the onshore winds thus enhancing coastal pollution.

The sea breeze fronts propagate inland during the course of the afternoon and can affect regions up to 50km from the coast in mid latitudes, although their effects are generally felt the most within 10 km of the coast, depending on topography, degree of urbanism and large scale circulation. Sea breeze impinging on higher terrain or against large scale offshore flow, or converging sea breeze from both sides of a peninsula (e.g. Florida, Cornwall) generate a zone of convergence and may cause clouds, convective rain showers and thunderstorms. Sea breeze circulation is common over Southern England and East Anglia (Simpson, 1994; Damato et al, 2006).

It is also important to note that a sea-breeze circulation is in essence a three-dimensional circulation, with an onshore surface flow and offshore flow aloft during the day. Many steady-state models, using only a single 10m surface wind as input data, will fail to represent a sea-breeze circulation, because of a non representation of the vertical structure and the spatial variability.

Levy et al. (2008) attempted to quantify the coastal recirculation effect on air pollutants by using a five year dataset of 30-minute averages of meteorological and air pollution data at 29 monitoring stations located at three air sheds along the Israeli coastline of the East Mediterranean Sea and at inland locations. In their study the highest concentrations of primary pollutants such as NO_x and SO_2 were measured whenever the daily average wind speeds were low, and particularly under poor ventilation conditions (i.e. low wind speeds and high recirculation factor). A recirculation factor is indeed objectively quantified in this paper (Levy et al, 2008) based on the ratio of L (discrete integral quantities of “resultant transport distance”) and S (net vector displacement, “wind run”), while for other pollutants such as O_3, higher values were found for both high and low recirculation factors. High ozone concentrations may have possibly resulted from long-range transport or coastal recirculation.

Leach and Patrinos (1992) showed how coastal circulations could influence local flow fields and deposition patterns. The synoptic conditions expected in the area they studied (Washington DC, USA) were modified by the coastal circulations and the expected deposition pattern was shifted to the north of the city.

A study by Speer and Leslie (2000) studied the atmospheric conditions which led to smoke pollution over the Sydney area due to a prescribed fire located north of the city centre. The very stable atmosphere, due to a formation of surface temperature inversion, associated with a succession of sea breezes and land breezes created an inter-regional circulation of smoke and accumulation of particulate pollution in the eastern part of the metropolitan area. Light wind conditions are usually good conditions for prescribed burning to avoid wild spreading of fires, but the inter-regional recirculation in this case created a major pollution impact over the Sydney area.

2.2.2 Anabatic and Katabatic Winds

Katabatic wind is the generic term for downslope winds flowing from high elevations of mountains, plateaus and hills down their slopes to the valleys or planes below.

Katabatic winds can be locally driven by cooling denser air flowing down the slope by gravity. For example cooling during night-time can cause a katabatic flow in the early morning when the cold air produced at high elevation starts flowing and accelerating down the topography. Katabatic flows slumping down from uplands may be funnelled and strengthened by the landscape and are then known as mountain gap wind, mountain breeze or drainage wind. Mountain breezes are part of a local wind system. When the mountainside is heated by the sun, the mountain breeze breaks down, reverse and blow upslope. These winds are known as valley winds or anabatic winds.

The gentler katabatic flow down hill slopes can produce frost hollows. This may occur after a dry, clear and cold night when cold air drains down neighbouring slopes into a localized pocket from which it is slow (or unable) to escape. Rickmansworth, a very well know frost hollow in UK, recorded the largest daily temperature range in England when, on 29th August 1936, the temperature climbed from 1.1°C at dawn to 24.9°C within 9 hours! Other well-known frost hollows in the UK are the Welsh Marches, the Glens of Scotland, the Pennine Valleys, the Vale of Evesham, Shrewsbury and Redhill. Frosts are often seen here earlier in the autumn and later in the spring than on the surrounding higher land (BBC). Note that where cold air can pool, dense gases may also be able to accumulate.

Katabatic winds can also occur on the lee side of a mountain situated in the path of a depression. Föhn type winds (such as the Chinook or the Helm wind) are known for their rapid temperature rise, their desiccating effect and the rapid disappearance of snow cover. These winds are typically found in the lee of large mountain ranges but can also occur in the lee of less marked mountains such as the Helm-winds in the Cross Fell Range in Cumbria.

2.2.3 Mountain and Valley Winds

2.2.3.1 Flow Recirculation in Mountain-Valley Wind Systems

In mountainous terrain, night-time downslope flows converge into valleys and make their way downstream, bringing (usually) fresh air downslope. Conversely during the day, upslope flow carries air up-valley. This thermally-driven terrain-controlled circulation reverses twice a day, soon after sunset and sunrise.

Baumbach and Vogt (1999) studied pollution trends in Freiburg, a town located in a valley in the Black Forest area in Germany. They showed how the mountain-valley wind system brings relatively unpolluted air masses from the Black Forest to the town at night and in the early morning hours during summer high-pressure weather conditions. They also showed how this cleaning effect fails to work during stable weather conditions with low wind speeds. Indeed under those conditions, it is the polluted air masses which have flowed into the Black Forest during the day that are transported back to Freiburg with the mountain wind in the evening and at night. Under these stable conditions, no fresh air comes in and recirculation increases the pollutant concentrations over the town. This example shows the importance for the model to have a memory of the previous several time steps to be able to predict correctly the pollutant concentrations over the town during such non-steady-state situations.

2.2.3.2 Shading Effects

Differential shading in complex terrain areas creates gradients of temperature and modifies local mountain-valley flow patterns (e.g. Maffeis et al, 2001). Differential shading effect also arises over flat terrain between cloudy and clear sky areas.

Additionally, differential shading creates non-homogenous dispersion, with unstable conditions developing earlier in the morning on the east-facing slopes and lasting later on the west-facing slopes. Finally, photochemical reactions develop differently in cloudy and sunny areas, most notably affecting ozone production.

2.2.4 Radiation Temperature Inversions

A temperature inversion may take place near the surface or higher in the troposphere. The latter type of inversion, aptly called subsidence inversion, is caused by large scale subsidence and was discussed in Section 2.1.1.2. A surface or radiation inversion is the result of surface cooling due to radiative heat loss during the night under clear sky conditions with low wind speed (and hence low mechanical turbulence). This type of inversion usually dissipates as the sun heats the ground in the morning, which can then lead to what is known as inversion-breakup fumigation (see Section 2.2.6.1).

However sometimes the morning inversion fails to break up at dawn and remains for several days, trapping pollutants near the ground and creating acute pollution episodes. These long inversion episodes are typically associated with a stationary high pressure area, weak winds near the surface, high humidity and persistent fog. The air being thermally stable, there is very little vertical motion, thus cold and very humid air generates fog at night. The fog, in these cold conditions, persists during the day, which prevents the solar radiation breaking the inversion layer by warming the ground in the morning and dispersing the pollution. These situations are considered non-steady-state because despite the winds being calm and the vertical turbulence in the layer quite stable, a build-up of material emitted under the inversion happens, leading to a non-steady-state situation. Steady-state models having no memory of the previous hour can not in theory simulate a build-up of material each hour starts with a clean air domain. Developers of some advanced steady-state models such as ADMS-4 have incorporated an additional module, for special treatment of ‘calm’ wind conditions, to palliate to this limitation (ADMS-4 User guide, 2010).

In the last two centuries, the worst air pollution episodes in London have occurred under radiative inversion conditions as described above, characterised by calm winds and cool, humid air, which developed into fog near the ground. Pollutants emitted in the stable layer under the inversion were mixed with fog to create what is called “smog” (combination of smoke and fog). For instance in 1952, the smoke from coal burning got trapped under a five-day temperature inversion creating a deadly “black fog”. Similar incidents were reported in London in 1956 and 1962. Each of them claimed from 700 to 4000 lives. A similar deadly event occurred in 1930 in the Meuse Valley (France) when pollution became trapped in a narrow valley. In the United States as well, such events have been recorded with a temperature inversion lasting six-days in Donora, Pennsylvania in 1948 and a three-day temperature inversion over Thanksgiving week-end in 1966, in New-York City, causing illness and deaths of a number of people.

However, in certain situations, such meteorological conditions might be beneficial and not detrimental to air quality. For example, on December 11, 2005, an accidental explosion generated massive fires at a Hertfordshire oil depot (Buncefield fuel depot), but thanks to a strong inversion layer, the hot elevated plume never reached the ground. Instead the lofted plume and its products were trapped at a moderate altitude. The plume emitted from the fire pierced the thin wintertime boundary layer and was injected into the free troposphere at higher altitudes. No high PM_{10} concentrations were recorded at any of the many air quality stations in the vicinity of the explosion. In addition, the study of the health impact of this fire, performed by Hoek et al, 2007, shows that acute public health impact was relatively small. At the time of the explosion, local temperatures were around freezing, wind-speeds were low and anti-cyclonic conditions prevailed (Jones et al., 2006). On the 2nd day after the explosion, the strength of the fires diminished and the plume became more narrowly defined because of an increase in wind speed and a more consistent wind direction from the North East. Ground-level concentrations of a range of pollutants remained low to moderate over local, regional and national scales. The conditions of the event (high plume buoyancy and favourable meteorological conditions) meant that the plume was trapped aloft with minimal mixing to the ground (Targa et al., 2006). If such an event had happened into a well developed summer boundary layer, the outcome would have been very different and might have caused severe air quality degradation owing to PM_{10} (Vautard et al, 2007).

2.2.5 Urban Heat Islands

The urban heat island effect is due to the presence of a city big enough to generate an atmospheric temperature larger than its surroundings, owing to anthropogenic heat release and heat storage in concrete buildings, roads and roofs. It creates meteorological changes in the area and impacts the atmospheric dispersion of pollution. Heat island magnitudes are largest under calm and clear weather conditions, often found during anti-cyclonic weather (Wilby, 2003), and especially at night, when it is common to observe neutral or unstable conditions over a city and very stable conditions outside the urban area. The location of the thermal maximum has been observed to change with wind direction (Graves et al, 2001).

Not only do heat islands create spatial heterogeneity in the meteorological conditions but they also create transient “city breeze” circulations which are by nature non-steady-state (they develop overnight and abate by mid-morning).

Nielson-Gammon (2000) compared two model simulations over the Houston metropolitan area. One simulation included the city of Houston; in the second simulation, the city characteristics were removed and changed to rural characteristics. During the afternoon, winds and temperature patterns were the same in the two simulations over most of the domain except where the city was located. Over Houston, the temperature was up to 2 degrees Celsius warmer than in the surrounding rural area. A convergent “city” breeze developed over the metropolitan area in the simulation including Houston and not in the other simulation.

Heat island circulation and sea breeze can compound each other's effects in dramatic fashion, such as in Chicago, where the sea breeze from the Great Lakes clashes with the city breeze over Chicago forming a cold front of sorts and causing severe thunderstorms over the city (WGN Weather, 2008).

Similar heat island effects can be found over lakes where industrial facilities discharge the water used for cooling purposes.

2.2.6 Fumigation

2.2.6.1 Inversion-Breakup Fumigation

Pollutants emitted above a radiation temperature inversion are trapped in the upper layer of the atmosphere during the night and isolated from the ground. As the solar radiation heats the ground in the morning the temperature inversion layer breaks up and turbulence within the now deeper boundary layer brings the pollutants aloft down to the ground, in a process called 'inversion breakup fumigation'.

Zhang and Rao (1999) have shown that ozone and its precursors trapped aloft in the nocturnal residual layer can influence ground-level ozone concentrations on the following morning as the surface-based inversion starts to break up. Figure 7, extracted from Zhang and Rao (1999), shows vertical temperature and ozone profiles in New Haven, Connecticut, at 5am and 3pm, and in Manassas, Virginia, at 8am and 12pm. Ozone has higher concentrations aloft (above the inversion layer) in the early morning hours and the concentrations become larger on the ground in the afternoon after the inversion layer breaks up. A one-dimensional model simulation supports their observation that the vertical mixing process contributes significantly to the ozone build-up at ground level in the morning as the mixing layer starts to grow rapidly. When the top of the mixing layer reaches the ozone-rich layer aloft, high ozone concentrations are brought down into the mixing layer, rapidly increasing the ground-level ozone concentrations.

A study by Anquetin et al (1999) shows the build-up and destruction of the inversion layer in a valley. It shows the influence of the season on the building of the inversion layer at night and its destruction in the morning.

Other experiments studying fumigation effects were conducted in complex terrain areas. For example, the tracer experiment in the Brush Creek Valley in Colorado, US in July-August 1982 described by Whiteman (1989) and Orgill (1989) shows the effect of morning fumigation when the convective boundary layer grows upward from the heated valley slopes. This experiment, staged in a mountain-valley area, was strongly dependent on the asymmetry of sun exposure of the sides of the valley at sunrise. Muller and Whiteman (1988) ran similar experiments to study the breakup of a temperature inversion layer in Switzerland's Dischma Valley on August 11, 1980. Allwine et al. (1992) also studied the formation of a cold air pool in a valley, isolating pollutant from the ground at night, followed by fumigation in the morning.

2.2.6.2 *Shoreline Fumigation*

An illustration of shoreline fumigation is provided in Figure 8, extracted from Luhar and Sawford (1995). Shoreline (or coastal) fumigation occurs when a plume emitted at the coast above the marine boundary layer is blown onshore by the sea breeze and encounters a growing land Thermal Internal Boundary Layer, known as the TIBL. The plume is initially travelling over land in a nearly non-turbulent unmodified onshore flow with little diffusion. Subsequently, the plume is intercepted by a growing turbulent boundary layer and undergoes rapid vertical mixing. This can lead to high ground-level concentrations of pollutants.

Sawford et al (1996) studied shoreline fumigation under sea breeze conditions in the vicinity of the Kwinana power station in Western Australia. This region is Western Australia's main site for heavy industry with most installations concentrated on a strip of land extending about 10 km along the shoreline. Wind, turbulence and temperature structure of the boundary layer, surface radiation temperature over both land and sea, as well as concentrations of CO_2, O_3, NO_2 and NOx, were measured during the 9-day experiment (between January 26 and February 6, 1995), dubbed the Kwinana Coastal Fumigation Study. Temperature and wind structure at the coast and further inland were measured at approximately two-hour intervals. Plume sections were measured near the Kwinana Power Station stacks and up to about 5km downwind. This study showed that during most of the 9 days the onshore flow was neutrally stratified but essentially non-turbulent. The growth of the TIBL in this neutral layer was rapid and limited by inertial rather than buoyancy forces. It was also found that there was a significant wind direction shear between the 10-m level at Hope Valley (approximately 3km inland) and the bulk of the TIBL. This shear had an important effect on the location of the ground level impacts of the plumes. The plume from the lower of the two stacks studied was clearly observed to fumigate regularly throughout the study within a few kilometres from the stack, while the plume from the taller stack generally stayed above the TIBL for the periods observed.

The results from this study show the importance for a model to be able to represent temporal changes in meteorological parameters for an accurate prediction of local pollutant concentrations. It also underlines the importance of properly modelling pollutant accumulation. Data from the experiment might be available from Australian Commonwealth Scientific and Industrial Research Organization (CSIRO) for model validation purposes.

2.3 Combination of Changing Meteorological Conditions

A combination of the characteristics responsible for local change in meteorological conditions such as an urban area, located along the coast on one side and surrounded by mountains on the other side, can amplify the development of changing meteorological conditions. Indeed, many studies have shown that for coastal cities which are heavily populated and surrounded by mountains, a combination of coastal recirculation, topographical settings, Urban Heat Island, and large-scale synoptic flow, has a strong effect on air quality.

For instance, studies all over the Mediterranean Basin show that during the summer season the combined effects of the sea breeze, local topography and synoptic flow often results in elevated levels of both primary and secondary pollutants (i.e. Clappier et al., 2000 study of the city of Athens in Greece).

Other meteorological combinations are also discussed in this section.

2.3.1 Combination of Land-Sea Breeze and Subsidence Inversion

On days where the Pacific anticyclone situated off the California coast creates large scale subsidence and a temperature inversion over Los Angeles (LA), severe smog develops over the city. When the afternoon sea breeze then kicks in, polluted air from LA spreads towards many inland locations, up to 60km away from the town. The air is warm enough to prevent cloud formation and plenty of sunshine is available to promote photochemical reactions (Simpson, 1994). The meteorological conditions leading to such events are clear skies and strong solar radiation, a critical balance between synoptic forcing and local sea breeze systems, which enhance pollution recirculation. In the 1940s, LA, California, became one of the first cities in the U.S. to experience severe air pollution problems because of this type of situation. The most serious pollution events in LA are related to land- and sea-breeze reversal, which gives a mechanism for a complete layer of polluted air to be maintained at high concentration and returned to the same locality 24 hours later (Simpson, 1994).

2.3.2 Combination of Land-Sea Breeze, Mountain-Valley Winds and Subsidence Inversion

Chang et al. (1989) and Kurita et al. (1990) studied a combination of land/sea breeze, and mountain/valley winds under synoptic-scale high pressure which created steady onshore winds, strong thermal low and subsidence inversions, associated with high ozone concentrations reaching inland mountainous regions (150km downwind of Tokyo) in the early evening. In the city, maximum concentration peaked in the early afternoon, when the sea breeze circulation developed. Under the combination of the above conditions, city polluted air was brought inland towards the mountain areas as shown in Figures 9 and 10, extracted from Kurita et al. (1990).

2.3.3 Combination of Land-Sea Breeze, Drainage Flows and Temperature Inversion on Strong Anti-cyclone Days

The city of Hobart, Australia is located in a well-defined valley with the Derwent Estuary running through its axis. The valley axis is mostly aligned in a north-west to south-east orientation with Mt. Wellington, the dominant topographical feature, approximately seven kilometres to the south-west of Hobart. Hobart is documented to have two dominant mesoscale wind flows, namely a sea breeze and katabatic drainage flows. The dominant daytime wind regime during winter is a drainage flow down the valley axis referred to as the “mountain wind”. This wind increases in strength and frequency with distance down the valley. The mountain wind is fed by down-slope drainage winds (katabatics flows) flowing off the valley walls to the Estuary. Light winds are generally associated with the mountain wind. High concentrations of particulate pollution in Hobart are frequently associated with the occurrence of highly stable atmospheric conditions and light winds that are unable to disperse pollutants. These conditions are linked to the passing of an anti-cyclone. Clear skies during calm wind events at night result in the cooling of air in the upper slopes of the Derwent Valley. The air slowly drains down the valley (katabatic winds) entraining pollutants within them. As a result, relatively high pollutant concentrations are likely to be found in topographic hollows and basins, and on low-lying land often located near the coast.

3 ATMOSPHERIC DISPERSION APPLICATIONS

Atmospheric dispersion modelling is used to estimate the concentration of pollutants at various distances and directions from a source for a wide variety of applications, ranging from accidental releases to regulatory permitting applications. A number of examples are discussed in this section.

3.1 Accidental Releases

In the event of a release of toxic material into the atmosphere, an accurate forecast of the initial plume transport and dispersion must be obtained within minutes to hours of the accident. Ground level air concentrations, and also deposition and irradiation from radioactive plume if relevant should be simulated by atmospheric dispersion models. Depending on the size and conditions of the release, it can also develop into a large scale event. And so, accurate modelling of the initial release and dispersion is not only necessary for short-term local predictions but also for longer term forecasts. Simulations of long-range transport trajectory over days to weeks then need to be provided.

Simple models such as steady-state models and simple meteorology may be enough for reporting results soon after the accident and at a short distance from the source if in conditions of non-zero wind speed and non changing meteorological conditions. However, in near-field situations where changing meteorological conditions occur often enough and have an impact on decision making in case of an accident, it would be more appropriate to use a non-steady-state model run with simple meteorology. In such situation, there may be insufficient time to identify whether or not the situation is steady-state and to decide which model to use. Long-range transports of pollution are certainly not steady-state situations and forecasting of such transport requires the use of models which remember the previous hour concentration and can take into account any change in meteorological conditions between the source and the receptors. As the release duration and extent of dispersion increase, the ability to simulate the spatial and temporal variability of meteorological conditions becomes more important. The transition between necessity for simple modelling and more complex modelling is not so easy to determine. For distances smaller than a transitional distance and timescales of a few hours, simple modelling could be adequate if the local spatial conditions are not too complex and the meteorological conditions are slowly changing. As soon as the distance travelled by the pollutant becomes greater than the transitional distance and the timescale increases to days or more, the use of a non-steady-state model that can simulate spatially and temporally varying meteorological and dispersion conditions would be recommended to adequately represent the path and duration of exposure. A question remains about how to determine the transitional distance where the conditions change from steady-state to non-steady-state. This distance varies and needs to be defined for each specific application. The computation of a steady-state index, dependant on local meteorological characteristics, as described in section 4.4.3 may help to evaluate the transitional distance.

The most important issue about an accidental release is the availability of meteorological datasets at the moment of the accident and in the following hours. If the accidental release happened at an industrial site, monitoring and forecasting of wind speed, wind direction and other meteorological parameters may be recorded on site and can be used for the dispersion modelling simulation. However, accidental releases may occur during the transportation of a pollutant and in such a scenario, meteorological data is more difficult to acquire in a relatively short timescales following the accident and required for dispersion simulation.

Examples of short-range and long-range accidental releases are discussed below.

3.1.1 Short Range Accidental Releases

Because of the acute health risks, especially in the close vicinity and immediate aftermath of an accident, it is paramount to model the location of the plume and duration of exposure within a degree of accuracy required to put an emergency response strategy together, including warning and evacuation of population and the safe dispatch of emergency teams. Employees working in facilities with possible risk of chemical releases are usually trained to stay upwind when evacuating for such an accident. Short oscillation of the wind direction, pooling and stagnation, structure confinement and building downwash, can all make the difference between life and death, for highly toxic releases. It is therefore important to have a high resolution grid and high resolution meteorology, both in time and space, to be able to predict the location of the plume and determine exposure accurately.

An accurate description of the release is also essential. Depending on the circumstances, the dispersion model should be able to handle time-varying emissions, buoyant, neutral, or dense gas releases, point sources, jet-like sources, area sources or volume sources.

Modelling of physical and chemical reactions may also be required, such as evaporation, dual phase releases, and chemical transformations.

3.1.1.1 Toxic Spills

Toxic spills of Ammonia, HF, or H_2S are examples of industrial accidental spill releases. Several modelling phases need to be addressed from the spillage itself (spills, dual-phase jet, etc), to the short-range dispersion (heavy gas dispersion when the gas is concentrated, neutral gas dispersion once the heavy gas is diluted enough), and possibly up to long-range dispersion.

Hydrogen fluoride is used by some refineries in the manufacture of unleaded gasoline. Amoco Corporation arranged with the Department of Energy to spill

1000 gallons in two tests at the HazMat Spill Center (formally called the National Spill Test Facility) near Mercury, Nevada, to study HF dispersion after a spill. This series of gas dispersion experiments are known as the Goldfish test series (Blewitt et al, 1987a, b), which can be and have been used for model validation (e.g. Hanna et al, 1991).

An actual accidental HF spill took place in 1987 at Marathon Corporation refinery at Texas City, Texas. A crane accidentally dropped equipment on top of a pressurized tank containing liquid HF. An estimated 36,000 lbs of hydrogen fluoride evaporated and escaped from the tank during the first hour after the top pipes were sheared plus perhaps another 4000 lbs during the second hour before the tank reached atmospheric pressure and was isolated. The fluoride plume was described as 2 to 3 miles long and 0.5 to 1 mile wide. The wind was from the SE at 5 to 10mph. Technical details on effects of community exposure to hydrogen fluoride during the Texas incident have been published in a paper by Dayal et al. in 1992.

Ammonia is one of the most commonly transported hazardous materials, especially in agricultural areas where it is used as an important fertilizer. It is also a common refrigerant and is frequently used in industrial areas. Ammonia is usually produced from natural gas, so it is also found in large quantities near petroleum producing areas. It is shipped in ships and barges, rail tank cars, and tanker trucks. Anhydrous ammonia is normally shipped in liquefied form (refrigerated on barges, pressurized on smaller carriers) and immediately vaporizes when lost. The major hazards associated with ammonia are from the toxic effects on breathing and caustic burns caused by vapour, liquid, or solutions. In spite of its low molecular weight relative to that of air, ammonia is able to form denser-than-air mixtures on release to the atmosphere. Depending on process conditions, ammonia can be released as a neutrally buoyant gas or as a heavier-than-air vapour cloud. A single phase release of gaseous ammonia may occur when ammonia is released from a small hole in a container where ammonia is stored in gaseous form. A two-phase release occurs when ammonia escapes from a pressurized vessel (where ammonia is stored in its liquefied form). In this case, the release cloud is typically denser than air. Besides storage conditions, meteorological conditions also affect how ammonia clouds evolve. Ammonia vapour can be readily advected and dispersed after an accidental release. However, during stable conditions, an ammonia cloud can linger around the spill area for quite a long time. Dispersion modelling of such an accident must be capable of handling both stagnant and windy conditions. Additionally, anhydrous ammonia may cause water vapour to condense and disperse as a dense aerosol close to the ground. Ground temperature and relative humidity are also factors influencing how ammonia disperses.

On January 18, 1992, a train derailment which sent a cloud of anhydrous ammonia over Minot, North Dakota, killed one man, sent part of a rail car slamming into a house and forced dozens of people to hospital with breathing problems. The air temperature was about 5^{o}F below zero. The cold temperature and a lack of wind made the gas linger in the area (according to Bismark Tribunes News Stories). Reports of such ammonia spills abound in the news literature, however the challenge for model testing purposes is to identify a case with good meteorological and monitoring data. Cawton et al (2009) analyzed ambient air-sampling data following accidental releases of ammonia. Although their focus was indoor, valuable information might be accessible in their dataset.

3.1.1.2 Emergency Flares

Emergency flaring occurs during operational shutdown caused by defective operations or planned maintenance in the oil and gas industry. During such mishaps, large quantities of gas are flared for hours or even days on end, with potentially significant releases of SO_2 and unburnt H_2S.

Short-term impact in the vicinity of the flare (hours, within 10km) can be addressed with steady-state modelling as long as the steady-state model can also address rainfall and vertical wind shear (the latter because the source is very buoyant and plume rise is significant). Obviously if micrometeorological properties vary sharply in the vicinity of the flare (for example for a close offshore or coastal location, or for a release occurring near sunrise), non-steady-state type of modelling may be required to address the changing meteorological conditions.

For longer range transport of SO_2 and H_2S from a lengthy emergency flaring situation, the meteorological conditions are unlikely to remain constant along the plume trajectory and non-steady-state modelling has to be performed.

3.1.1.3 Gas Blow-By and Pipe Ruptures

When a control valve fails and is stuck wide open, for example in a Water-Oil-Separation Plant (WOSEP) at an oil-gas production facility, high-pressure gas could find its way out of a tank or pipe. The jet-type accidental release is usually short-lived (from a few minutes up to a couple of hours, during which operators shut-down or isolate the defective system). Although the release itself is time-varying, the impacts are generally confined within short distances (hundreds of metres) and meteorological conditions are unlikely to vary between the source and receptors, unless the rupture occurs in a cluttered built-up area. Therefore, meteorological conditions are typically steady-state during blow-by and pipe rupture accidents.

3.1.2 Long Range Accidental Releases

Accidental releases can have lasting airborne effects, of the order of days or even weeks, long after the sources have stopped emitting and as long as the pollutant is airborne. In those cases, long-term meteorological conditions have to be considered. Because of the long distances and short and long timescales involved, steady-state modelling is not an option for long-range dispersion modelling of accidental releases.

A typical example of lasting airborne accidental release is the Chernobyl nuclear accident, with radioactive material reaching the upper troposphere and being transported far away for a long period of time (weeks to months). Superposed on the general trend of decreasing fallout with increasing distance, are more local incidence patterns reflecting weather conditions. Rainfall, thunderstorms, or any subsidence event can bring material down to the ground far away from the site of the original accident, and long after the initial incident has occurred.

The Chernobyl explosion is such an example, with radioactive material spewed all over Europe, and radioactive rainfall occurring, notably, in the UK. During the two day passage of the Chernobyl cloud over the UK, on May 2-3, 1986, heavy thunderstorms and rainfall were the major factors affecting local deposition of radioactive material, especially radioactive Cesium (Cs-137). A survey undertaken by the Institute of Terrestrial Ecology (ITE) recorded levels of ^{137}Cs deposition on vegetation ranging from less than 10 Bq m^{-2} in parts of the Midlands and Southern England to over 1,000 Bq m^{-2} in many Western upland areas more affected by rainfall at the time (Allen, 1986). A simple steady-state model is not adequate to correctly predict the amount of radioactive Cesium that deposited over the UK from the Chernobyl cloud on May 2-3, 1986.

Other examples of long-range transports are described in section 3.8 related to natural sources releases.

3.2 Risk Assessment

Risk assessment studies are performed by facilities conducting potentially dangerous operations in order to design safety zones around those operations. Safety perimeters are based on accident type, released chemicals, failure frequency, and meteorology. Risk Assessment can also include the quantification of risk associated with accidental releases of short duration. Risk assessment impact results are used for input to emergency planning. Contrary to actual accidental releases, which were discussed in the previous section, risk assessment modelling does not require the knowledge of a particular plume path at a specific time. Air dispersion modelling studies are implemented to estimate the frequency of the worst case scenario and at which distance from the source the highest peak concentrations may happen. Both air concentrations and flux depositions at different timescales are a concern.

Steady-state modelling might appear to be conservative and sufficient for risk assessments since the distance from the source to where maximum concentrations occur is usually within 10 kilometres. But for short time-scale accidents, it is important to remember that worst-case scenarios might involve non-steady-state situations, such as stagnant conditions followed by fumigation. Stagnant conditions are steady-state per se, but the accumulation of pollutant they create and subsequent flushing by changing meteorological conditions cannot be handled by steady-state dispersion models.

An analysis of the local physical characteristics of the area around the sources and the frequency of certain type of meteorological conditions may be required. Local changes in meteorology can be linked to the worst-case impact. Such conditions need to be identified and their frequency evaluated. If they can lead to worst-case scenarios and are frequent enough, a steady-state model may not be appropriate for risk assessment analysis in the near-field of such a site.

3.3 Odour Modelling

Odours are the most important environmental issue in implementing wastewater treatment and bio-solid management facilities, although many other industrial and agricultural processes also cause odour nuisance. The time scale for odour can be as short as 0.1 to 1 second and is usually in the sub-hourly time scale. The averaging time specified in odour legislation is location specific: for example, it is one hour in Massachusetts, US, Europe and UK, ten seconds in Hong Kong and one second in Australia.

One important requirement for modelling odours is the capability to take potential stagnation and accumulation into account (for example during calm wind conditions), as well as compounding factors such as recirculation and building downwash. Causality effects and spatial and temporal variability are also important factors.

Whether steady-state modelling is adequate or not depends on the timescale involved and the type of odour application. Indeed if the odour modelling is performed to assess the potential for odour nuisance in the vicinity of a malodorous facility, steady-state modelling might be adequate, provided recirculation or stagnation is not an issue. If however odour modelling is performed to assess a specific complaint and the necessary high-frequency meteorological data is available, one might have to actually model high frequency meandering of the malodorous plumes, with a puff, particle, or non-steady-state CFD model.

Odour modelling being in general a near-field application can in some cases also have a long-range impact as it is demonstrated in Smethurst et al. (2010) paper. In this paper, the authors tried to understand why a number of odour complaints were registered over a large area of East UK on the morning of April 18, 2008. The area of concern was much larger than a possible local impact. Their conclusions described possible large scale spreading of agricultural slurry over Belgium, the Netherlands and north Germany during low wind speed conditions followed by brisk easterly winds bringing the stagnated air towards the east coast of England. Steady-state atmospheric dispersion models would not be able to reproduce such situations of possible long range transport, which includes stagnation followed by changing wind characteristics becoming stronger and unidirectional.

An extensive review of available dispersion models to assess odours was published by the National Environmental Research Institute in Denmark (Olesen et al, 2005). The authors discuss Gaussian plume models, Lagrangian particle models, and CFD models. They mention but fail to discuss puff models. The report also describes a selection of available datasets for model validation.

3.4 Regulatory Impact Assessment

These assessments are designed to evaluate the impact of future sources for permitting purposes or to evaluate potential upgrades to reduce the excessive impact of existing sources. Source apportionment analyses, worst-case scenario evaluation, engineering design and cost-benefit analyses can also be conducted in such studies. The regulatory control assessments are usually carried out for continuous releases. The impact assessments focus on peak concentrations (or nth percentile) for timescales varying from sub-hourly to annual averages. Consideration of planned short duration releases such as reactor blow down events or abnormal discharges may also be required.

Regulatory assessment can be required for long-range transport or near-field impact. Steady-state modelling is often sufficient for short range impact, although not always if the area of interest either experiences many calm periods or if it includes a physical boundary affecting micrometeorology, such as a coastline or a valley. The United States Environmental Protection Agency (US EPA) recommends using AERMOD for near-field impact assessment, however, in certain more complex situations a non-steady-state model such as the Lagrangian puff dispersion model CALPUFF may need to be used to better represent the situation. Long range transport usually requires non-steady-state modelling. The distance from the source where the transport becomes non-steady-state and a long-range application is usually site specific and needs to be evaluated beforehand.

Regulatory impact assessments of routine nuclear discharges, an example of regulatory assessment application, are commonly simulated using simple Gaussian plume models for annual average concentration estimations. Studies by Lutman et al. (2004) compared the impact results of such applications from a steady-state model (the Gaussian plume model NRPB-R91, Clarke R.H. 1979) and a non-steady-state (the Lagrangian particle based model NAME, Maryon R.H. et al., 1999). Both concentrations of radiative pollutants and flux depositions of pollutants were evaluated. Statistical meteorology rather than temporal meteorology was used as input into the models. One of the conclusions of the study was that the difference between the annual average concentrations for the two models was within the accuracy of the models themselves. And since the results of the Gaussian plume model were larger than the results of the Lagrangian particle model at a distance larger than 200km, it was concluded that simple Gaussian plume model associated with statistical meteorology can be accepted for such applications. The question that can be raised is whether the concentrations results from the Gaussian plume model may be too conservative or not but observations were not available for comparison. While considering deposition fluxes, the simple Gaussian plume model was not considered adequate to model wet deposition fluxes. The modelling results of wet depositions with the steady-state model were much smaller than when the Lagrangian particle model was used.

This illustrates that simple steady-state models and/or statistical meteorology have been used for long-range impact assessment on long-term timescales such as annual averaged concentration estimates. Considering the distance between the source and the receptors, the situation is clearly non-steady-state. In this case, statistical meteorology is used to palliate the steady-state characteristics of the models. However, regular practices for long-range applications at short-term and long-term timescales have been gradually changed to the use of non-steady-state models associated with sequential meteorology.

For near-field impact of a regulatory impact assessment over short timescales, the use of simple steady-state models can be questioned. Some complex flow situations such as sea breeze, mountain/valley breeze, fumigation or stagnation followed by front or fumigation can lead to peak concentrations in the near-field of a source and cannot be accurately modelled by a simple steady-state model. Model evaluation studies are needed to quantify the amplitude of the error on concentrations and flux depositions if simple steady-state models are used instead of non-steady-state models in such non-steady-state situations. The availability of datasets for such studies is discussed in section 5.

3.5 Operational Real-Time and Forecast Modelling

The user of atmospheric dispersion models for such application is interested in a conservative estimate of the impact of a facility in the vicinity of the sources in the following 24 to 48 hours to avoid violating health, safety or regulatory standards. If the pollution forecast approaches or exceeds a regulatory standard, the system should raise an alert, predict impacts for alternative operational scenarios, and help in the decision making to switch to less polluting operations. An example of such a system using ETA Analysis and CALPUFF modelling is described in Robe et al., 2002.

As for all short range applications, steady-state modelling should be adequate as long as there is no potential for recirculation, stagnation or fumigation. Additionally if the terrain is complex, the model, be it steady-state or non steady-state, should be capable of modelling terrain-induced circulations. For long-range forecast modelling application, non-steady-state models are recommended.

3.6 Planning Studies

Examples of such studies are land use planning to minimize population exposure to pollutants, design and optimization of monitoring networks, or selection of a site for implementing a new facility. The important outputs required from these studies are the concentrations and spatial distribution of the pollutants, the maximum distance from the source where the pollutant concentrations can violate standard thresholds for averaging periods ranging from sub-hourly to annual time scales, and the frequency of exceedances. In the modelling, the user needs to take into account the important geophysical features that can mitigate or enhance the impact (such as bodies of water, forests, heat islands due to urban city centres, etc).

As far as dispersion modelling is concerned, planning and permitting studies are rather similar.

3.7 Cumulative Impact Assessments

Cumulative impact assessments look at the combined impact of several sources, sometimes several hundred sources. For instance, in the United States, the National Ambient Air Quality Standards (NAAQS) are cumulative standards, not single source standards. Therefore background source contributions can be important and sometimes critical for NAAQS compliance demonstrations. It requires modelling of all background sources in the vicinity of the source of concern to estimate its compliance with the NAAQS.

If the pollutant of interest is a passive tracer, each source can be modelled separately. If however chemistry is important, all the sources have to be modelled together, with a model that can handle all of them as well as relevant chemistry, which is a rather restrictive requirement.

Whether steady-state modelling is adequate or not once again depends on the distance of interest and averaging time. Unless all the sources and receptors pertaining to the cumulative impact assessment experience identical weather, a model that can deal with non rectilinear trajectories is required and straight steady-state Gaussian plume models are not adequate. Moreover if the modelling domain is so large that pollutants cannot reach the receptors of interest before meteorological conditions change, non steady-state modelling is also required.

3.8 Natural Sources

Other atmospheric dispersion applications are developed for monitoring or forecasting natural sources emissions such as volcanic eruption, accidental and prescribed fires or even regional sand transport.

3.8.1 Volcanic Eruption

The first example of these applications is a volcanic eruption spawning ash way up into the stratosphere, with particulate matter circling the globe for months after the eruption, allowing the potential for contamination to last for a very long time, with deep convective events and large-scale subsidence areas responsible for bringing the impact to the surface sometimes months and thousands of miles away from the volcano.

Additionally minor eruptions and volcanic smoke are a constant threat to aircraft passing in the vicinity of volcanoes and the dispersion of ash in the lower and middle troposphere needs to be constantly and accurately predicted. The Particulate Matter (PM) plumes very much depend on the sporadic release (definitely a non-steady-state source) and the weather which is affected by mesoscale meteorology, terrain-related waves, and thermals. This type of non steady-state dispersion application had a direct impact in the UK and Europe in spring 2010, when British and European air space was closed for up to 10 consecutive days to aircraft because of the potential presence of volcanic ash (containing highly abrasive dust particles) dangerous for aviation (BBC, 15th April 2010). This situation arose as a consequence of the explosive activity from the Eyjafjallajokull volcano in Iceland (with ash ejected to a height of between 20,000 and 30,000 ft at times), the meteorological anticyclone system centred west of the British Isles, and the associated North-West winds advecting the Icelandic ash towards Europe (Met office, 2010). Other areas of the world with active volcanoes such as Sicily, Indonesia and Alaska need monitoring and volcanic ash pathways forecast to potentially divert aircraft flying over these areas.

3.8.2 Fires (Accidental or Prescribed)

Every year, square kilometres of forest burn in many parts of the world (Asia, North America, Russia, Europe, etc.) either on purpose or accidentally. For example, forest burn in Borneo, emitted smoke and ash all over South East Asia. Kuala Lumpur, experienced Borneo-fire-related haze during the month of August, when the large scale atmospheric circulation directs the ash plumes across the South China Sea. Morning inversion compounds the problem, with serious consequences for health and visibility (e.g. Afroz et al, 2003).

Another example of large scale fires and long range dispersion applications are the oil well fires in Iraq during the first Gulf War. Those fires were started on purpose but accidental well blow-outs often get ignited and result in fires lasting several days. Owing to the duration of the fires, meteorological conditions do tend to change during the course of the fires. Moreover the blow-out itself is not a steady-state release, with explosive and gaseous releases often preceding the ignition and subsequent fire. An example of such a blow-out occurred at the Ocean Odyssey Platform on the UK Continental Shelf, on September 22, 1998. Other offshore drilling fires include the Piper Alpha disaster on July 6, 1988.

Prescribed fires are usually started during adequate meteorological conditions so they do not bring any disruption in the vicinity areas and do not spread out of control. Low wind speeds conditions and low levels of turbulence are required to avoid such spreading. Speer and Leslie (2000) showed how a change in meteorological conditions during a prescribed fire can affect the local population and its activity. They studied an air pollution episode during the period 12-14 April 1997. This generic example of a stationary high-pressure ridge with its axis over the New South Wales coast just north of Sydney produced very light winds at low levels over the Sydney metropolitan area and aided the formation of surface temperature inversions, associated with a succession of humid sea breezes and land breezes. These meteorological conditions concentrated the smoke of a prescribed burn just north of Sydney in the eastern part of Sydney metropolitan area. The hazardous smog formation was suddenly transported south-west over a major highway disrupting the local traffic. It was induced synoptically by a change in wind direction that transported smoke and fog to the south west.

Another prescribed fire which may have affected population at a long distance from the source is discussed by Witham (2008) where she described how biomass burning in Ukraine in March 2007 may have led to elevated PM10 over much of the UK.

3.8.3 Sand Transport over long distances

Sand transport towards nearby cities, as it was illustrated for Mexico City in section 2.1.2.4, is another example of a natural source of pollutant dispersion that may require modelling. Such transport can be local but may also spread over very long distance. Long range transport of sand from the desert of Gobi or the Sahara has been shown to impact Beijing city and cities all over Europe, respectively. For instance, during the period 23-24 January 2008, eight sites in the UK measured levels of PM10 concentrations at air pollution index 7 (high) or above, and two of these sites also went on to record very high pollution at index 10. The cause of this PM10 particulate episode was observed to have been long range transport of dust as a result of sandstorms in Africa with a possible but unlikely contribution from African forest fires (Cook et al, 2008).

4 STEADY-STATE VERSUS NON-STEADY-STATE DISPERSION MODELLING

4.1 Steady-State Conditions

Non-steady-state models should be required to simulate dispersion applications occurring when meteorological conditions change significantly during the time it takes for pollutants to travel from source to receptor. However, steady-state models are sometimes used to model non-steady-state situations and the results of such modelling are appropriate in some specific situations. So, it is important to accurately define steady-state modelling conditions and steady-state model characteristics to be able to evaluate the suitability of steady-state models for modelling non-steady-state conditions.

Steady-state modelling conditions can be summarized as follows:

a Conditions do not change over time:
  - Over the time period needed for the plume to reach each receptor, the meteorological conditions are assumed to be constant
  - Source characteristics, including emission rates, exit temperature and exit velocity are constant

b Each hour is separate and independent of previous hours:
  - No memory of pollutant location or emissions from previous hours are required

c Meteorological conditions are constant within the modelling domain, which is true for most steady-state models, some having the capability to deal with varying terrain by modelling linear flow around complex terrain.
  - Spatially constant meteorological variables: wind speed & direction, mixing height, temperature, humidity, and precipitation
  - Spatially constant turbulence variables: Surface friction velocity (u*), convective velocity scale (w*), Monin-Obukhov length (L), all related to surface characteristics.

Although not strictly part of the Eulerian definition of steady-state conditions, the conditions in (c) are true for most steady-state dispersion models (see section 4.4.2 for a discussion of Eulerian vs. Lagrangian steady-state).

Steady-state models are appropriate for modelling pollution impact at mesoscale distances from a continuous-release source as long as the land characteristics are spatially constant between the source, the receptors and the meteorological stations involved in the modelling, and as long as the flow remains non complex. It is difficult to determine the exact distance from the source when the conditions become non-steady-state. It is dependent on the source characteristics, land surface conditions and meteorology. The steady-state index described in section 4.4.3 may help to determine how far from the source steady-state conditions are still valid.

Variability in meteorological conditions may not always be reproduced correctly with some steady-state models because of the nature of their characteristics as described above. For instance, steady-state models which represent plumes as straight lines to infinity are not able to represent curved trajectories. They are also unable to represent time of travel, which may have an impact when the wind speed varies. The combination of the two limitations can bring pollutant toward a receptor where the plume may not have reached. A study developed by the Atmospheric Study Group (ASG) at Earth Tech, Inc. for CALPUFF training to illustrate discrepancies between steady-state models and non-steady-state models, displays a 24h average footprint of SO_2 concentration (Figure 11) from hourly continuous emissions simulated with a steady-state model on the left (ISC) and with a non-steady-state model on the right (CALPUFF). The same meteorological data, in the form of a single surface station, is imported in the two models. CALPUFF outputs, like ISCs, are computed using single point meteorology. Figure 11 shows how the trajectories are extending to infinity at each hour on the left while the trajectories are following the variations in wind speed and wind direction on the right. The comparison of the two footprints shows a larger maximum 24h average impact for the steady-state model. The main impact for the two models is located in the North to East-South East side of the source. However, the steady-state model impact is covering also areas on the North West side and South side of the source. While the North West side and South West side of the source is never reached with the non-steady-state model.

Since in steady-state models all time steps are independent, no accumulation of pollutants can be simulated. Pollutant accumulation above the top of the planetary boundary layer before morning fumigation or pollution accumulation in a calm wind area before a sudden change in wind direction and intensity are situations that a steady-state model fails to simulate correctly. Similarly coastal fumigation associated with an onshore breeze and a change of mixing height between a coastal source and an inland receptor requires non-steady-state modelling.

Changes of land characteristics can induce changes in turbulence and create situations where pollutant concentrations are depleted by dry deposition or become more diluted. For instance, ground concentrations tend to be higher over smoother surfaces while rougher surfaces increase turbulence and help a polluted cloud to dissipate thus decreasing ground concentrations.

4.2 Time scales

Based on the examples discussed in the previous chapters, Table 1 summarizes timescales associated with changing weather conditions and various dispersion applications. This table highlights under which meteorological circumstances a specific dispersion application may need to be modelled with a non-steady-state dispersion model. The key is whether the meteorological conditions changed during the time it took for the pollutant to travel from its source to the receptors of interest: this could be a matter of minutes, hours, days or even weeks, depending on the application and the relative position of sources and receptors.

However, depending on the averaging time of interest for the application and the frequency of non-steady-state conditions, steady-state models may be appropriate for modelling non-steady-state situations. For instance, individual high impact events, such as fumigation and calm wind conditions, usually do not contribute too much to annual averages, unless the frequency of this type of event is dominant over all the year. Therefore, steady-state models are usually acceptable to compute annual averages at receptors close enough to the source for the pollutant to reach them within a time step or within the time scale of typical weather events in the area, and provided the trajectories to the receptors are straight line (for most steady-state dispersion models). For annual average impact at distances from a source which can no longer be considered steady-state, the use of steady-state models might be questioned. A study by Lutman et al. (2004) shows that annual averages of steady-state model results were conservative when compared to non-steady-state model ones at distances from the source of 200km or more, but the comparison was showing opposite results for impact at distances between 100 and 200km. For this application, statistical meteorological data was used for steady-state modelling. A discussion between statistical and sequential meteorology is tackled in section 5.1

When short-time scale averages are the focus of a study, the circumstances leading to the highest impact have first to be analysed. Indeed, even if simple steady-state models give usually a conservative estimate of concentration impact, in some specific situations it may not be conservative. If those circumstances involve accumulation, recirculation, or changing meteorological conditions along the trajectories towards the receptors, a non-steady-state model is required. If however the highest impacts are linked to specific meteorological conditions (e.g. very stable hours, high wind speeds, etc…) not involving accumulation or recirculation, a steady-state model should be able to capture the peaks. For applications, when the pollutant path is of concern, steady-state models need to be used with caution since they are unable to simulate curved trajectories. Figures 12 and 13, two individual time steps, hour 9 and hour 4, respectively, taken from the study developed by ASG, Earth Tech, Inc for CALPUFF Training (section 4.1) illustrate the discrepancies between simulations with steady-state models or non-steady-state models in curved trajectories situations due to changes in wind speed and direction. Figure 12 displays a higher peak for the steady-state model simulation and a curved trajectory for the non-steady-state model while Figure 13 displays a higher peak for the non-steady-state model simulation and a different location of impact than for the steady-state model simulation.

4.3 Non-Steady-State situations where use of a steady-state model can become an issue

The applications described in Section 3 can be classified by the nature of the outcome at the sensitive receptors. For some applications such as long-range and short-range accidental releases, odour modelling and forecasting, the pollutant path and its concentration along the path are crucial and need to be simulated accurately. On the other hand for risk assessment or regulatory impact assessment for permitting purposes or planning purposes, the maximum peak concentrations, worst-case scenarios and the frequency of peak concentrations are the most important.

For applications where the pollutant path is important, using a steady-state model when a change in wind speed or wind direction occurs between the source and the receptor has potential to overpredict or underpredict the pollutant concentrations at the receptors. For accidental release applications, it might result in incorrect emergency response decisions. For odour modelling and source apportionment, it might result in a misinterpretation of the source of the pollutant. For forecasting, it might result in giving wrong information to the public or making wrong operational decisions at industrial sites.

The main meteorological parameters whose changes can affect the pollutant concentrations or pollutant path include the wind direction, wind speed, vertical wind shear, turbulence or stability classes, mixing height or temperature gradient and precipitation. Table 2 links changes in meteorological parameters with changing weather situations, potential impact on receptors, and atmospheric dispersion applications which can possibly be the most affected by these changes.

Wind shifts affect plume trajectories and consequently the concentration footprints. Significant changes in wind direction along the plume path such as those associated with the passage of a front (warm or cold), thunderstorms or squall lines, or any air recirculation such as land-sea breeze, and mountain-valley winds, cannot be simulated with a straight line plume, a characteristic shared by most steady-state dispersion models. Using a straight line model under such changing circumstances might lead to large discrepancies with observations or simulations carried out with a path-following model such as Lagrangian puff model or Lagrangian particle model. Long-range accidental release, odour modelling, real-time operational, and forecast applications are affected by significant changes in wind direction. The longer the range the more likely the plume encounters a shift in wind direction along its trajectory.

A change in wind speed transports the material and the peak concentrations to a potentially different distance from the source than if the wind stays constant. A change of wind speed also affects mechanical turbulence resulting in changes in the dilution of the material within the toxic cloud. For example, pollutant concentrations accumulated during calm wind conditions can affect sensitive receptor areas when the wind suddenly increases and carries the polluted air over the sensitive area. Applications such as accidental release, odour modelling and real-time operational modelling are sensitive to such changes in wind speed. Smethurst et al. (2010) show how some odour modelling complaints over the UK could be linked to long range transport of material after a period of stagnation conditions.

Vertical wind shear can affect the path and the pollutant concentration. The surface wind may not be representative of the wind at the tip of a stack or at the height where the buoyant source is released. In case of accidental release or emergency response for instance, it is important to incorporate the vertical resolution of wind speed and wind direction and sometime the three dimensional resolution of wind speed and direction. For example, in the case of a source located at a coastal site and subject to sea breeze circulation.

Changes in turbulence conditions on the path between the source and the receptors may be significant, like for instance, if the material is transported from a rural area to an urban area (or vice versa). As the roughness length changes, so do the turbulence level and hence the plume dilution. Smaller roughness lengths (rural area) induce less turbulence, less dilution, and therefore usually larger concentrations than larger roughness lengths (urban or forested areas). As many steady-state models assume uniform roughness length over the domain, they fail to properly model impacts across non-uniform areas, either underestimating or overestimating ground concentrations depending on the choice made for that single uniform roughness length. This can affect accidental release applications as well as regulatory impact assessment applications for any averaging period. Modelling efforts might be required to quantify the impact of simulating an area with uniform versus non-uniform roughness length where it is relevant.

Changes in mixing height can have a strong effect on pollutant concentrations at the ground. Such a change can create a sudden increase in ground concentrations when the polluted air masses are mixed to the ground by a growing turbulent boundary layer. Examples of such situations are inversion break-up and shoreline fumigation effects. On the other hand, on a clear sky night, radiative cooling of the ground generates a stable surface layer, capped by a thermal inversion layer, which can trap pollutants either above the inversion layer, with beneficial consequences as in the Buncefield Depot Fire, or below the inversion layer, with harmful consequences as in smog events. These mixing height changes play an important role in the atmospheric dispersion of pollutants. They have a great impact on applications either sensitive to the amount of pollutant at receptors, or focusing on worst-case scenarios and peak short-term concentrations. Applications such as odour modelling, risk assessment, regulatory impact assessment on short-term time scale, operational real-time modelling and forecast modelling are affected by change in mixing height between the source and the receptor. These situations are considered non-steady-state and require non-steady-state atmospheric dispersion models for adequate modelling.

Changes in precipitation have a strong impact on pollution deposition and removal of material from the atmosphere. Precipitation can take multiple forms depending on air temperature and path travelled by the air parcel. The most common forms are rain, snow or hail. Gaseous pollutants are scavenged by dissolution into cloud droplets and precipitation. Particulate pollutants are removed by both in-cloud scavenging (rainout) and below-cloud scavenging (washout). Different types of precipitation have different impacts on pollution. For instance, liquid precipitation can scavenge gas while frozen precipitation usually does not. For modelling purposes, empirically-based scavenging coefficient methods are used. For advanced models, the scavenging of a pollutant depends on the precipitation rate, the nature of precipitation and the characteristics of the pollutant itself (e.g. solubility and reactivity). Acid rains are an example of the consequences of pollution being trapped in clouds and washout with rain on vegetation. An accurate simulation of acid rains is affected by temporal changes in precipitation conditions along the path of the pollutants. However, if the precipitation occurs close to the source or is homogeneous on a determined period, steady-state model would be able to reproduce wet deposition fluxes in the vicinity of the source. Long-range accidental release event such as Chernobyl is an example where the transport of pollution in an air mass was suddenly drained out in an area because of a sudden change in meteorological conditions. Such event, characterized by a long-range impact (hundred kilometres from the source), cannot be modelled with a steady-state model. The model needs to be able to simulate removal of materials along the path between the source and receptors and represents accurately the path of the pollutant up to the area of concern. A steady-state model, characterized by a straight-line trajectory and no memory of the previous hour cannot simulate hourly or daily wet deposition fluxes of pollutant at such distance from a source accurately. A long-range impact study of routine nuclear annual discharged performed by Lutman et al. (2004) shows that even on long-term averages a steady-state model would fail to reproduce the annual average of wet deposition fluxes. The wet deposition fluxes computed by R91 were much lower than the wet deposition fluxes computed by the NAME model, a Lagrangian particle model. The modelling results were not compared to any observations since the latter were not available but nevertheless, estimates obtained with the steady-state model were much too low to seem reliable. Although homogeneous rainfall can be dealt with by steady-state models, rainfall occurring sporadically or locally between the sources and receptors requires non-steady-state models for proper modelling.

Changes in land use characteristics along the path of a pollutant can affect the dry deposition fluxes if modelled. For instance, in simple Gaussian plume models, the dry deposition flux is a function of the ground concentration and deposition velocity specific to the pollutant. The reduction in air concentration is spread uniformly across the plume by modifying the original source term for example in the PLUME model (Jones, 1981), while in Lagrangian puff models such as CALPUFF (Scire et al, 1996) the reduction due to deposition velocity or wet scavenging is applied differently at every time step along the path, depending on local conditions of landuse and meteorological information.

4.4 Further Discussions

4.4.1 Calm Wind Conditions

Although the focus of this work has been on temporal variations of meteorological conditions, it is important to stress that non-steady-state models might be required even when the meteorological conditions are apparently non-changing. One such instance is during calm wind conditions.

During calm wind conditions the hourly averaged meteorology is steady but the dispersion model has to be non-steady-state to account for pollutant accumulation during calm wind conditions if they last more than one hour. If a steady-state model is used during those calm hours, peak concentrations are likely to be underestimated. This certainly affects short-term averages but might not impact longer term averages unless calm wind frequency in the area of interest is high. Both the frequency of calm wind conditions and the length of the calm wind periods are important to be considered. One hour of calm wind may be of little impact both for short-term and long-term averages but several consecutive hours can lead to high pollutant concentration. If these several hours of calm wind happen frequently enough during the year, long-term averages may also be affected.

Steady-state Gaussian plume models cannot handle zero wind speeds because of their formulation: both plume rise and horizontal plume spread are inversely proportional to the wind speed. Calm wind hours are therefore either removed from the computation, or a minimum wind speed is applied. For instance, ADMS-Urban and ADMS-road set a minimum wind speed of 0.75 m/s. While the US EPA type models (AERMOD, ISC) assume that all wind speeds recorded as between 0.5 – 1 m/s are treated as 1m/s. For wind speeds less than 0.5 m/s, a number of rules are applied either to ignore these hours if short period of calm wind is recorded or to apply a minimum wind speed value if long period of calm wind is measured. A lot of research is still being carried out to circumvent that limitation. For instance, an option has been added to the latest version of ADMS, ADMS-4 to simulate calm wind conditions (CERC, 2010).

It is also worth noting that calm hourly-average winds do not imply the absence of any motion, but rather they imply high frequency sub-hourly multi-directional wind shifts. While the sub-hourly shifts may not matter much for long scale dispersion, as long as the overall plume growth and accumulation are accounted for, the sub-hourly shifts may be important for short-range transport of toxic or malodorous compounds. For instance, the short range peak impact may be at a different spatial location if hourly averaged or sub-hourly averaged meteorological conditions are used in the modelling. Barclay (2008) showed how modelled surface concentrations can change quite substantially if averaged hourly winds are used rather than 6-minute averaged winds (Figure 14). Additionally Figure 15 (from Barclay, 2008) shows a different spatial footprint and a large increase in turbulence variability if high resolution real time turbulence parameters are used rather than model-computed turbulence parameters.

4.4.2 Spatial Variability

Both spatial and temporal changes in meteorological conditions can significantly impact pollutant dispersion. Although spatially varying meteorological conditions within a modelling domain can be a priori steady-state, it is important to note that most common steady-state dispersion models:

a are straight Gaussian plume models
b assume uniform land use (i.e. uniform dispersion properties)
c assume uniform rainfall (if any),
d even sometimes assume flat terrain

Therefore, even in steady-state meteorological conditions, many steady-state models are not appropriate to simulate dispersion over non-uniform domains.

Furthermore, steady-state meteorology from an Eulerian point of view (i.e. constant meteorological conditions at a given location in the domain) does not imply steady-state meteorology in the Lagrangian sense (constant meteorological conditions along the pollutant's trajectory between the source and the receptors). From the pollutant's point of view as it travels from the source to a given receptor, spatially inhomogeneous meteorological conditions do mean temporally varying meteorological conditions.

The limitations of straight plume models are illustrated in Figures 16 and 17 (from Scire et al, 2009). Figure 16 depicts the situation of multiple sources, two of which are on the coastal side of a sea breeze front, and one of which is on the land side of the front. Very different wind directions in the two areas make it impossible for a steady-state model to predict regional impact accurately in this situation. Figure 17 depicts the situation of sources located within a curved valley. Terrain channelling of the flow requires the use of a non-steady-state model that can deal with spatial variability of the flow or a steady-state model which has an option to include a contour following module such as ADMS.

So whether the meteorological conditions are changing rapidly at a given location within the domain or whether they are changing spatially within the domain, non-steady-state dispersion modelling is required to accurately model the changes in dispersion when the outcome is on a short-term timescale.

Meteorological events only spatially affect a dispersion application if the spatial changes are within the modelling domain. In Figure 16, a steady-state model could be used if only one source had to be considered (i.e. no cumulative impact) or if the three sources were on the same side of the sea-breeze front for the averaging period of interest. Similarly in the Figure 17 example, straight plume model at the source called INKOM is appropriate as long as the receptors are located in the same valley segment, i.e. no further away than 6 km during easterly wind conditions but only up to 1 km in westerly flow.

4.4.3 Steady-state Index

For modelling purposes, an analysis may need to be performed to identify how often during the modelling period and over which areas conditions can be characterized as steady-state. Scire (2009) introduced the notion of a steady-state index (SSI) to help assess the “steady-state status” of a given application. He further suggested basing the SSI on spatial and temporal variability of three factors within the modelling domain: dilution, as measured by wind speed, advection, as characterized by wind direction, and dispersion, based on stability class. If any of those parameters varied significantly at any time or place between the source and the receptors, non-steady-state conditions applied and if they happen often enough during the period analysed a non-steady-state model should be used.

4.4.4 Source characteristics

Source characteristics, such as release height, exit velocity and exit temperature, may affect pollutant trajectories and dispersion, and how they are affected by changing meteorological conditions. Ground or non-buoyant source impacts are typically shorter range than impacts from elevated or buoyant sources, and consequently not as likely to encounter varying meteorological conditions.

Varying emissions or intermittent emissions are non-steady-state of course but beyond the scope of this review.

5 ATMOSPHERIC DISPERSION MODELS AND EVALUATION DATASET

5.1 Choice of Atmospheric Dispersion Models

One of the aims of this review is to determine how current atmospheric dispersion models account for changing meteorology and how this affects modelling results. The number of atmospheric dispersion models has increased enormously over the years. Some models are more widely used for regulatory applications while others are usually designed for risk assessment or accidental release modelling. It is not the purpose of the review to describe the models themselves but rather to acknowledge how different the models are in terms of incorporating meteorological observations and computing dispersion parameters, and to find which ones are more suitable to simulate changing meteorological conditions in each application

A few factors need to be taken into account to decide which model is adequate for a given application. When the worst-case condition is the requested outcome for the application, steady-state models used to be associated with statistical meteorology to fulfil this requirement. Non-steady-state models are usually more sophisticated and include more complex parameterisations. They also require more meteorological data input, more computer time and more expertise. Whether the extra effort required to gather both data and expertise is really necessary depends on the application. More specifically it depends on the type of application, the locations of the sources and receptors, source types, complexity and variability of the meteorology, desired accuracy of the results (i.e. highly accurate versus conservatism) and averaging time.

Meteorological Data Availability

Most countries in the world have their own network of meteorological measurements of surface and vertical profile parameters. The coverage of such observations can be sparse in some areas and not available at the appropriate time step, however atmospheric dispersion modelling needs meteorological input without missing time steps and recorded as close as possible to the local area of interest. Before the wide distribution of prognostic meteorological mesoscale models output were available, alternative methods to palliate missing data were developed, such as the use of statistical meteorological data to track the frequency of the worst meteorological conditions for dispersion. Nowadays, in the UK and most developed parts of the world, data availability is no longer an issue since mesoscale forecast systems are routinely run by the local meteorological offices, providing both forecast and past analyses. From those, single point meteorology time series or three-dimensional meteorological fields can be extracted and imported into steady-state models or top-of-the-line Lagrangian and Eulerian dispersion models, respectively. Hourly mesoscale datasets are also computed by a number of organisations for most places in the world at 12km and 4km resolution. Such multi-year global datasets are available for instance from the UK Met Office or from the MM5 dataset developed by TRC (TRC-ASG website: http://www.src.com/mm5/MM5_Main_Page.html).

Computer time

Current IT advances make computer time no longer an issue, except possibly for real-time emergency response applications. A full year and four sources can be simulated on a domain of 200 x 200 grid points with a Lagrangian dispersion model such as CALPUFF in a few hours. By increasing the number of grid cells, and the number of receptors, the computer time will augment accordingly.

Expertise

Expertise is required for developing and applying any dispersion models. However once a system is set-up, non-experts can usually perform further applications and interpret them. The experience of users, the air quality ambient standards, and the consistency with previous studies have to be taken into account in the choice of a model.

Accuracy versus Conservatism

Simple steady-state dispersion models are commonly used in the UK for regulatory impact assessment for all averaging periods. The argument to justify their use even for non-steady-state application is that they are simple, easy to use and usually provide a conservative estimate of concentration impact. Although, this is a general statement and the users need to be aware that in certain meteorological conditions the opposite can be true and steady-state models can simulate lower concentrations than non-steady-state models. As shown in section 4.1 and 4.2, steady-state models do not always give the highest concentrations for short-term averaging and in the vicinity of the source. For long-term averaging (such as annual averages), the long-range study performed by Lutman et al. (2004) showed that the simple steady-state model results always exceeded the Lagrangian model concentrations at very long distance (over 200km away from the source). However in that study, steady-state model results did not simulate the highest impacts at distances between 100 and 200km from the source.

Averaging Time

A requirement for either peak hourly concentrations or annual averages impacts on the model choice. Applications looking at short-time averages must be able to represent extreme, often non-steady-state events. The importance of isolated extreme events decreases when long-term averages are of interest.

5.2 Types of Dispersion Models

All existing models cannot be described. In this section, categories of models are differentiated from one another by how much and which type of meteorological information goes into the model and how meteorological and dispersion parameters are computed internally. The meteorological parameters that are important for dispersion modelling include directly measured parameters such as wind (speed and direction), temperature, and precipitation. Other parameters such as dispersion coefficient and mixing height can either be provided as observations or computed internally using surface friction velocity (u*), convective velocity scale (w*), Monin-Obukhov length, solar radiation, sensible and latent heat fluxes, stability classes and ground characteristics such as albedo and roughness length. Most of the atmospheric dispersion models mentioned in this section are listed on the online European Model Documentation System (MDS) which can be consulted at http://pandora.meng.auth.gr/mds/strquery.php?wholedb for reference and for a more complete description of these models.

5.2.1 Simple Gaussian Plume Models

The simplest dispersion models are the simple Gaussian plume models. A few examples of this type of model are R91, SCREEN, and PLUME (part of PCCREAM suite of models). Most of these types of models can use sequential observed meteorology from one local station or statistical meteorological data computed from a number of years of sequential local meteorology and use defined stability tables for turbulence estimation. A number of tables have been developed for various parts of the world and different applications. An example of such table is the 60% Category D stability class distribution, which is a good assumption for meteorological conditions over the UK when considering long-term averaged impacts (Clarke R.H., 1979).

5.2.2 Advanced Gaussian Plume Models

The more complex Gaussian plume models such as ISC3, OLM, BLP, AERMOD and ADMS can also input statistical meteorology but more frequently incorporate one-dimensional sequential meteorological information. This information can be direct observations from a meteorological station or output from a prognostic model. It can consist of a full vertical profile or just surface observations. Some other improvements that can be found in these models when compared to the simple Gaussian plume models are an increased knowledge of turbulence and diffusion in the planetary boundary layer, calculations on plume spread that are based on conditions occurring at the height of plume rather than at ground level and calculation of the vertical spread of pollutant by assuming it is non-gaussian. Dispersion coefficients are computed using micro-meteorological parameters such as surface friction velocity, Monin-Obukhov length, roughness length. The Pasquill-Gifford-Turner (PGT) dispersion curves are used for this purpose. These curves were developed using the Prairie Grasse experiment (Barad, 1958) and are more suitable for simulation in rural areas. AERMOD imports the surface roughness length and the Monin-Obukhov length values at the closest meteorological station available and computes spatially constant dispersion coefficients. ADMS imports Monin-Obukhov length, boundary layer height and the wind speed to estimate these coefficients. The atmospheric turbulence is simulated in those models by the computation of dispersion coefficients. The simple (section 5.2.1) and advanced Gaussian plume models are steady-state models in the sense that the meteorological parameters imported or computed are constant spatially within the domain for each hour and that the conditions remain unchanged on the pollutant path between the source and any receptor, no matter how far from the source they are located. Indeed, the assumptions for steady-state Gaussian plume models are constant condition within a time step (i.e. hour), straight-line trajectories, non-zero wind speed, no causality effect (do not account for travel time between the source and receptor) and no memory of the previous hour (each hour is separate and independent of previous hours). Some of the models such as ADMS have options to treat calm wind conditions, to adjust the flow to topography or to import a file with spatially varying roughness length and create spatially varying dispersion coefficients which makes the modelling somewhat non-steady-state. But these are only adaptations to the physical local conditions. The source of meteorology stays one-dimensional and the time independence of these models prevents them from being fully non-steady-state.

5.2.3 Lagrangian and Eulerian Models

The common characteristics of the third group of models are that they can input a three-dimensional dataset of meteorological information and are non-steady-state models. Within this group, the dispersion models can be divided into a few other categories: the Lagrangian puff models, such as CALPUFF, UDM or SCIPUFF, the Lagrangian particle models such as NAME, MicroSpray, part of model system MSS (Tinarelli et al., 1994, 2000), AUSTAL, and QUIC-plume, and the Eulerian models such as CMAQ, EMEP Unified Model, and CALGRID. Some other models such as TAPM, a hybrid Eulerian/Lagrangian or HYSPLIT (hybrid single-particle) also import three-dimensional meteorology. As for the advanced gaussian models, these models compute dispersion coefficients internally with various refined parameterizations using imported or evaluated micro-meteorological parameters. The variation in parameterization of the dispersion coefficients from one model to the other can induce discrepancies between modelling results but probably less important than importing three-dimensional meteorological data rather than one-dimensional meteorological data when the impact due to changes in meteorological conditions between the source and the receptors is the main concern. The non-steady-state models allow three-dimensional meteorology, spatial variability to winds, turbulence fields, precipitation and temperature. They allow variable and curved trajectories, spatial variability of terrain and landuse. They retain information from the previous hour, allow calm wind and low wind speed conditions and include causality effects.

Changes in wind direction and wind speed have probably the greatest impact on predicted concentrations at a given point. As described in section 4.3, wind direction is used to estimate the path trajectory of the pollutant while wind speed is used to determine plume dilution and plume rise downwind of the source, which affects the magnitude of and distance to the maximum ground level concentrations. Short-term averages are more sensitive to these changes and long-term averages less so. The issue is to determine how significantly results are affected. Gaussian plume models incorporate wind data information from a local point (varying temporally only), which is used for the entire domain. The plume extends downwind from the source to infinity. The winds are extracted at the source height for the more complex models and at ground level for the simple ones, and there is no memory of the previous time step. Each time step of the modelling starts with a "clean" footprint. Lagrangian puff models or Lagrangian particles models on the other hand incorporate three dimensional wind fields (varying spatially and temporally). The distance travelled by the pollutant in this case is determined by the wind speed. These models remember the previous hour modelled and the foot print resulting from the emission of the new time step is added to the previous time step footprint. These three characteristic discrepancies, (i) travel to infinity versus fixed finite travel distance, (ii) not remembering versus remembering the previous time step footprint and (iii) single point wind data versus three dimensional wind fields, have an effect on the location and the concentration of the highest peaks. Any applications which are sensitive to the exact location and the amount of pollutant predicted display large discrepancies when using a simple Gaussian plume model versus a non-steady-state Lagrangian puff or particle model. The shorter the time average impact the user is interested in, the stronger the discrepancies are.

5.2.4 Other Models

The Computational Fluid Dynamic (CFD) models (Code_Saturne (CFD RANS), FLUENT, MERCURE), which can more accurately resolve building structures, obstacles, and the flow around them can also be mentioned. Other models designed for accidental release of dense and toxic gases such as HGSYSTEM, SLAB, DEGADIS**,** GASTAR, PEAC-WMD software are usually simple dispersion models importing simple meteorological data. Some of these models, such as DEGADIS, use statistical type of meteorology and are unable to use sequential data.

5.3 Evaluation Datasets

Most field study databases which include all the information needed for model evaluation have in general been developed to improve atmospheric dispersion models. These field studies were usually designed to evaluate specific characteristics of models. Atmospheric dispersion datasets can be classified as follows:

- Dense gas and toxic gas accident release studies (like for example SMEDIS) – very short-term emissions, receptors extended up to 6-10 km from the source – dense gas or neutral gas.

- Turbulence / near-field tracer experiments - vicinity of the facility, flat terrain or simple terrain features – buoyant gas, continuous emissions, and receptors extended to 10-20 km, 50 km at most from the source.

- Long-range tracer experiments – short-term or continuous emission over a few days up to annual period of monitoring. – buoyant gas.

- Other sets of experiments are developed to study circulation of pollution in urban areas, looking at the effects of buildings on flow. For instance, wind tunnel experiments fall into this category.

5.3.1 Dense gas and toxic gas accidental release studies

A number of accidental release datasets were used by Hanna et al. (1993) to evaluate atmospheric dispersion models specialised for dense gas dispersion modelling. Only a limited number of accidental releases, where hazardous chemicals purposely released into the atmosphere for field experiments, have been carried out and even fewer have their test results in the public domain. One of them for example, called “Goldfish Test Series”, was conducted during the summer of 1986 by Amoco Oil Company and Lawrence Livermore National Laboratory at the Haz Mat Spill Test Centre. The tests consist of six anhydrous hydrofluoric acid releases. The results are presented in a paper by Blewitt et al., 1987. A constant discharge rate is maintained during the test. Receptors were placed on arcs at 300 metres, 1000 metres and 3000 metres downwind at a dry lake bed known as Frenchman Flat. Like most of the existing control hazardous chemicals released, the winds blow in a predictable direction and are more or less constant during the time of each series and the meteorological conditions correspond to a “D” atmospheric stability. This example demonstrates that the interest in the outcome of hazardous chemicals accidental release is the concentrations at a distance of less than ten kilometres and change in meteorological conditions have not yet been of strong interest. During these types of experiments, which are of a short duration, the wind speed and direction are usually more or less constant and the meteorological conditions are neutral or stable in the Pasquill-Gifford definition. The “Desert Tortoise” series of tests conducted in 1983, which released ammonia, are described in a report by Goldwire et al., 1985. This second example also displays no interest in changing meteorological conditions: the receptors were placed up to 5600 metres from the source, the wind was constant for each series and the stability classes were either neutral or stable. Only short-term change in meteorological conditions can have an effect on such an application. If the toxics do not stay in a dangerous phase for a period long enough, a change in meteorological conditions in the few seconds or hours following the accident in most cases would not make a significant difference if simulated by a steady-state or non-steady-state model. The peak is estimated by both steady-state and non-steady-state models with only small discrepancies between the two relative to the degree of model and input data uncertainty. However, if the toxics can stay in the atmosphere for a few hours to days and weeks with a concentration harmful to the human population or the environment, steady-state models can fail to predict the correct path and/or potential accumulation or deposition.

5.3.2 Near fields tracer experiments

A large number of near-field tracer experiments have been conducted over the years for evaluating the performance of atmospheric dispersion models. Three of them, developed on flat terrain areas, widely used for atmospheric dispersion models evaluation are Project Prairie Grass (1956), Kincaid (1980-1981) and Indianapolis (1985). Project Prairie Grass is a tracer experiment of SO_2 release in rural surrounding from a near ground level source. The sample concentrations are 10-minute samples at downwind distance from the source from 50 m to 800 m. Half of the samples were measured during daytime and half of them during nighttime (Barad, 1958; Haugen, 1959). The Kincaid is a tracer experiment conducted at Kincaid which involved a release from a 183 m stack with a buoyant plume rise over a flat terrain rural area. 171 experiments were conducted. Measurements were hourly for both near surface ambient concentration and meteorology. Receptors arcs ranged from 0.5 km to 50 km from the source. A large number of the measurements were recorded during the afternoon in spring and summer, period representative of daytime convective conditions (Bowne et al., 1983). The Indianapolis SF6 tracer experiment is a complex urban site experiment conducted at Indianapolis city. It involved a release from an 84 m stack with buoyant plume rise. Measurements of hourly near surface concentrations at a distance of 0.2 km to 12 km from the source and hourly meteorology (Murray and Bowne, 1988) were recorded. For more extensive description of these near-field tracer experimental datasets or get access to other similar datasets, the US EPA website (http://www.epa.gov/scram001/dispersion_prefrec.htm) and John Irvin's website (http://jsirwin.com/Tracer_Data.html) can be consulted. Most of these tracer experiments are near-field applications in flat terrain environment, assuming non changing meteorological conditions. However, some datasets, developed to test the limitation and refine steady-state models, may include some non-steady-state situations. One of these datasets is a tracer experiment called the Tracy Power Plant Experiment. The power plant is located on a flat plateau surrounded by terrain features. Tracer gas was released through the 91.4-m smokestack of an active power plant located near Reno in Nevada, US. Meteorological measurements from a 150-m tower are also available. Concentration monitoring in the surrounding terrain were done mostly during late evening and early morning hours. The complex terrain features and the development of morning inversion layer, which breaks up as the sun rise, create non-steady-state conditions that could be used for sensitivity testing steady-state models versus non-steady-state models in those conditions.

In the UK, a few near-field tracer experiments data sets are also available. For instance, three field tracer experiments (Technology And the Study of Atmospheric Dispersion in the Urban Environment) (http://urgent.nerc.ac.uk/Meetings/2001/Abstracts/simmonds.htm)) performed in Birmingham City, UK during 1999 and 2000 whose main goals were to test a new technique for measuring the tracer and also to provide the scientific community with a dataset for dispersion over spatial scales between 1 km and 10 km. However, experimental arrangement needed to be as simple as possible from the dispersion point of view. Near-neutral stability conditions and a wind speed of about 4-5 m/s were chosen in order to satisfy the requirements for the simplest meteorological conditions for the experiment. These studies do not fulfil any changing meteorological conditions either.

As we observed above, near-field studies have been deployed under a number of meteorological conditions (stable, neutral, convective, etc…) for testing the simulation of the early version of atmospheric dispersion models (mostly steady-state models), but very few data sets available consider the impact and evaluation of models under changing meteorological conditions. A possible explanation is that the first models were steady-state models and so the field studies were adapted to the characteristics of these models (indeed steady-state). More recent and more complex models with non-steady-state characteristics are also validated against these data sets and usually give a good performance.

5.3.3 Long-range tracer experiments

The best relevant field experiments which include changing meteorological conditions are the long-range Tracer Experiments. However these tracer experiments being conducted to study long-range pollution impacts usually have their ground concentrations measurements starting at a distance more than 50 km away from the source. At such a distance, steady-state models are usually not the preferred models to be used.

A large number of long-range experiments have been developed over the years, just a few are discussed here as examples. For instance, the European Tracer Experiment (ETEX) project consisted of two releases to atmosphere of tracers sampled for three days after the beginning of the emission using a sampling network spread over a large part of Europe. This experiment was performed to evaluate the performance of a large number of non-steady-state dispersion forecasting models. Another example of such an experiment is the European eXport of Precursors and Ozone by long-Range Transport (EXPORT). The primary objective of EXPORT was to characterise and quantify the photochemical air pollution formed over Europe and exported eastwards from Europe. The data held at BADC was collected during a co-ordinated three aircraft flying campaign in August 2000 based at Oberpfaffenhofen in Southern Germany. Measurements were made of many photochemical parameters including ozone, its precursors, other oxidants and both gas phase and particulate tracers in the air over Europe and that being transported eastwards out of Europe.

Five other long-range tracer experiments data can be accessed in the Data Archive of Tracer Experiments and Meteorology (DATEM). This archive provides an opportunity to link high quality modern meteorological data with the data from five long-range tracer experiments performed over the United States from 1974 to 1987: 1) ACURATE – the Atlantic Coast Unique Regional Atmospheric Tracer Experiment from 1982 and 1983, 2) ANATEX – the Across North America Tracer Experiment from 1987, 3) CAPTEX – the Cross Appalachian Tracer Experiment from 1983, 4) INEL74 – Idaho National Engineering Laboratory releases in 1974 and 5) OKC80 – a single tracer release from Oklahoma City in 1980. Currently, only longer range (hundreds to thousands of km downwind) experimental data are considered (Draxler et al. 2002). The U. S. National Center for Environmental Prediction (NCEP)/National Center for Atmospheric Research (NCAR) meteorological re-analysis using historical data (1958-1997) and analysis of the atmospheric state during this period have been enhanced with many sources of observations not available in real time for operations, provided by different countries and organizations. The measurements of concentrations vary from 12 to 24 hour averaged concentration over a period of 19 months (ACURATE- the Atlantic Coast Unique Regional Atmospheric Tracer Experiment from 1982 and 1983 (Heffter et al., 1984)) to 3 hour samplings on a period of a few days (OKC80 – a single tracer release from Oklahoma City in 1980 (Ferber et al, 1981)).

5.3.4 Urban Tracer Experiments

Experiments studying atmospheric dispersion in urban areas were developed and are available for roads and streets in many cities of the world. One example of these types of experiment is the DAPPLE (Dispersion of Air Pollution and Penetration into the Local Environment) project which has been deployed since 2003. Four field campaigns have been completed between 2003 and 2008 in and around the intersection of Marylebone Road and Gloucester Place in London. Such observational datasets are examples of local urban experiments developed to test the performance of urban dispersion models. By bringing together fieldwork, wind tunnel and computational simulations, it is expected to provide a better understanding of the physical processes affecting street and neighbourhood scale flows of air, traffic and people. However, for such experiments, the dates and timing are usually chosen with meteorological conditions as stationary as possible to be able to study the flow around buildings for certain wind directions for instance.

5.3.5 Other Experiments

In addition, experiments which involved meteorological measurements in non-steady-state situations are available and could be of interest for testing the performance of steady-state models versus non-steady-state models in changing meteorological conditions. These experiments provide only meteorological support for the sensitivity analyses. They can not be used for evaluating models since the sources characteristics and emissions are not provided and the pollutant concentrations are not monitored as it is in tracer experiments. They can nevertheless be used to compare the sensitivity of atmospheric dispersion models in non-steady-state meteorological situations. A couple of meteorological experiments are described below.

The Improved Air Quality Forecasting (ISB52) experiment, concentrating on studying meteorological flow parameters, was developed to gain a better understanding of air flow within the atmospheric boundary layer in the vicinity of an urban area by gathering three-dimensional air flow information using two identical Doppler lidar measurements. Field experiments were undertaken in March 2003 at Malvern and in July 2003 at RAF Northolt, West London, UK (Bozier et al., 2004, Davies et al., 2007). The March 2003 experiment during winter type conditions under an anti-cyclonic system recorded the effect of a temperature inversion at night, while the July 2003 experiment covered a wide range of meteorological conditions during summer varying from large scale anti-cyclonic systems to small scales features such as showers and thunderstorms. Comparisons with a couple of models regularly used in the UK such as the UK Met Office air quality forecasting model NAME or ADMS were performed.

A second set of meteorological data archived at the British Atmospheric Data Centre (BADC) is the surface meteorological data and high resolution radiosonde data from the Met Office's research site in Cardington, Bedfordshire. The dataset contains recorded surface measurements timed at 1, 10 and 30 minutes intervals. Wind is measured at 10 meter, 25 meter and 50 meter above the ground level. Some measurements performed at the Cardington research site on the period August, September and November 2005 were used for testing improvement methods of low speed wind simulation in TAPM model (Luhar, 2007). Low wind speed condition is not exactly changing meteorological condition but it is a situation non-steady-state plume models usually can not simulate (except if a specific option to treat cam wind conditions as in ADMS-4 was added) due to their non-steady-state assumptions.

5.4 Sensitivity Tests in Changing Meteorological Conditions

Potential discrepancies between steady-state model impacts versus non-steady-state model impacts for a number of applications have been discussed in the previous sections of this review. Although the discussions were mostly qualitative, it would be very useful to be able to quantify these differences to validate the choice of modelling with one type of model or the other. We are proposing a selection of tests to be performed in future work to fulfil this goal. A choice of datasets, models and type of tests are discussed below.

Datasets needed for a thorough atmospheric dispersion models evaluation in changing meteorological conditions require a complete set of information which includes local meteorological measurements during the event, accurate emission rates and measurements of concentrations and/or flux depositions at a number of receptors of interest. The meteorological and emission data are used as input into the model to be evaluated and measurements of concentrations and/or flux depositions compared to the model output simulations. However, unless a field experiment is specifically designed to study an event or certain characteristics of a model, all information required is not always available. Despite an extensive number of field experiments described in section 5.3, it appears that changing meteorological conditions are of concern mostly for long-range tracer field experiments. However, long-range tracer experiments are not suitable for evaluating steady-state models, especially at short-term time scales.

A few of the near-field experiments described in section 5.3 documents changes in meteorological conditions and represents non-steady-state situations. The Tracy experiment is an example including changing meteorological conditions which are the breakup of morning temperature inversions. The ISB52 field experiment studies atmospheric processes in an urban area and the Met Office's research site in Cardington, Bedfordshire could provide high resolution meteorological data for low wind speed conditions. These two latter datasets focus on measurements of meteorological parameters but provides neither source emission rates nor concentration measurements. A third field experiment, called the Kwinana coastal study, was developed in Western Australia to study shoreline fumigation under sea breeze conditions (Sawford et al., 1996). More information is provided in section 2.2.6 and meteorological data, emission releases and pollutant concentrations might be available from this study.

We propose to use some of these experiments to develop a matrix of sensitivity tests to compare steady-state models versus non-steady-state models results in a number of non-steady-state situations. If observed concentrations are not available, model simulations might be compared to one another for an estimation of the discrepancy between models.

Changes in meteorological conditions are either directly linked to the physical characteristics of the area of interest or can happen anywhere. A local analysis of the characteristics of the modelling domain should assess whether the area is subject to either coastal fumigation or land/sea breezes, for instance if there is the presence of a water body in the vicinity of the source or whether the area is subject to valley/mountain breezes if the location of interest is in a mountain area, etc... Local recirculation and pollutant accumulation can result from such situations however steady-state models cannot reproduce such phenomenon. In addition, or if the terrain of the local area is not complex, a number of questions still need to be raised. Some questions are more relevant if peak concentrations or worst-case scenarios are of concern, such as:

- What is the frequency of meteorological conditions leading to calm wind, morning fumigation, recirculation?
- Will the steady-state model always give the worst-case scenario or the highest peak?

If the exact path or exact location of a peak pollution event is of concern, one has to track the frequency of meteorological conditions leading to a change of path for the pollutant, such as front passage, the possibility of precipitation along the path, etc...

For any outcome, to determine the frequency of changing weather patterns is crucial if looking at long-term events since if the pattern is frequent enough it could have an impact on long-term averages. Figure 18 displays a diagram showing a proposed procedure to determine if a non-steady-state model may be needed for the application of interest. This diagram raises potential questions a user could ask and is not assumed to be exhaustive. Figure 18 shows the potential complexity of the situations and puts into perspective where sensitivity tests between steady-state and non-steady-state models could be relevant to quantify their discrepancies.

Five selected non-steady-state situations are proposed for testing the sensitivity of steady-state models and non-steady-state models and for comparing the results of the models with observations, when it is possible. Table 3 summarizes the selected sensitivity tests. Three different types of models are proposed to be tested: a simple gaussian plume model (such as SCREEN or PLUME), a more complex gaussian plume model (such as AERMOD or ADMS-4) and a Lagrangian model (such as NAME or CALPUFF).

One test is specific for studying the impact of local land characteristics on atmospheric dispersion. The Kwinana field experiment includes all the data necessary for evaluating models in shoreline fumigation effects situations. Results of all three models applied with this dataset are compared to observations for quantifying the discrepancies. A second test will look at low wind speed conditions and how significant the discrepancies could be whether such situation is simulated with a steady-state or non-steady-state model. Two of the tests could use the field study ISB52, which provides meteorological observations as a three-dimensional field. Data for a few of the changing meteorological conditions events described earlier in this review are available in the ISB52 experiment: temperature inversion and morning inversion break-up and passage of fronts with showers/thunderstorms. The timescale is a few hours to a few days in the near-field. We suggest studying an elevated source and a ground source and to look at the concentration impacts of these sources for receptors located at distances from 1 to 10km from the sources using all three types of models mentioned previously. A thorough comparison of the outputs of the models is proposed to quantify the significance of discrepancies. For the morning fumigation, the Tracy power plant datasets could be used as a second dataset for testing the sensitivity of the models in this type of meteorological situations.

The proposed procedure shown on Figure 18 diagram to determine whether steady-state models are suitable or not for a specific application could be tested in parallel to the sensitivity studies. The computation of a steady-state index is also proposed to document each test.

Note that the suggested sensitivity tests are subject to the acceptance from the authors to grant access to their datasets.

6 CONCLUSIONS

The short-term impact in the vicinity of an accidental release can be addressed with steady-state modelling if the meteorological conditions are not too complex and the impact is relatively close to the facility. If micro-meteorological properties vary sharply (such as at a close offshore or coastal location or for a release near sunrise), non-steady-state modelling may be required to address the complex changing meteorological conditions. For long-range transport, since the meteorological conditions are unlikely to remain constant along the plume trajectory, non-steady-state modelling must be performed.

For risk-assessment, the frequency of the worst-case scenario is of interest, so changing meteorological conditions that can lead to peak impacts of pollution are the most important to single out and determine their frequency on the path between source and receptors. Typical situations include local areas subject to land/sea breezes or mountain/valley flows, or other types of air flow recirculation but also shoreline fumigation, areas with frequent morning fumigations or frequent long periods of calm wind conditions. If any of these situations are simulated with steady-state models, results must be treated with caution, since the examples cited above are situations where steady-state models may predict lower peak concentrations than non-steady-state models.

For regulatory impact studies, highest peak concentrations are usually of primary interest for averaging periods varying from sub-hourly to annual. The distance from the source at which the highest peaks occur is also of interest. Steady-state models are acceptable in most near-field situations however if the characteristics of the area are complex and flow recirculation or alternative weather pattern, leading to pollutant accumulation are common in this area, the use of a non-steady-state model should be considered. For any long-range applications, steady-state models are not usually recommended.

In conclusion, each type of application needs to be treated differently, a number of questions need to be raised and local meteorological analysis is advised to determine the potential for substantial change in meteorological conditions which could have an impact on the outcome of the application. A number of external factors listed below such as availability of correct meteorology, CPU time, consistency with other studies, etc… may also need to be taken into account in the selection of the appropriate model.

Questions to be raised: Initially, the question relates to the outcome of the application. Is the pollutant path or the pollutant concentration at the receptors the main focus of the study? Secondly, the relation to the concentration itself is important. How accurate does the simulation of the concentration need to be? Are we looking for a conservative estimate or a concentration as exact as possible? Most steady-state models predict a straight line path so are adequate for potential constant wind speeds and direction conditions between the source and receptor. Steady-state model concentration predictions tend to be conservative estimates at a certain distance from the source. So, for more precise concentration estimates, the use of non-steady-state models may be a better choice.

Time Scale: Firstly, for annual average estimates of pollutant concentrations, using steady-state or non-steady-state models in changing meteorological conditions might not impact significantly on the results in the vicinity of the release. For a long range application, the choice of meteorological input might be important. The use of statistical meteorological data with steady-state models is more likely to give conservative results at long-range receptors. Whether this is what the regulatory agency and the industries are expecting for the application can be debated. Whether the results are conservative or not at any distance from the release with this type of modelling is still a question. Some experimental modelling may be necessary to evaluate such statements.

Over short time scales, changing meteorological conditions are more likely to have an impact on the outcome of the applications. Using steady-state models for long-range applications at this timescale is not recommended. For near-field application, a study of the local area is recommended to estimate if changing meteorological conditions are likely to occur with a high frequency and what changing meteorological conditions must be present to determine if such changes lead to accumulation of pollutant or deviate the pollutant from a straight trajectory. The outcome of the application for short timescales is thus also important information to have in mind. If the exact path of pollutants is of concern and changing meteorological conditions can divert the trajectory of the pollutant, steady-state models are likely not to be appropriate for such modelling. If the worst-case scenario or peak concentration for a specific averaging period is required, any changes in meteorological conditions leading to pollutant accumulation are not simulated correctly with a steady-state model.

Meteorological data availability: Each time a non-steady-state model may be potentially a more acceptable choice the user should determine if the meteorological data required is readily available and consider the effort needed to access this meteorological dataset. Nowadays, even if observed meteorological data are not available in the vicinity of the modelling domain, prognostic meteorological datasets can be accessed from a number of websites (like the Met Office, or TRC-ASG, etc…). With a grid resolution high enough, these datasets can have better world wide coverage than actual meteorological observations. The user needs to have the capability to evaluate the provided prognostic meteorological datasets or request a thorough evaluation from the provider to make sure it is reliable and adequate for the intended application.

Additional factors need to be taken into account when choosing between a steady-state model and a non-steady-state model.

CPU Time: The facility and rapidity to run the atmospheric dispersion model can be an important factor in the decision. There used to be a significant difference in CPU time between running simple steady-state models and complex non-steady-state models. With current computer capabilities, this is less of an issue.

Uncertainties: A number of uncertainties are inherent to model parameterisations, meteorological parameters, source characteristics, input data and concentration measurements. Discrepancies between models outputs and observations may need to be put in perspective with these potential uncertainties to identify their significance.

Type of Source: Elevated sources are more likely to be affected by change in meteorological conditions than ground sources. Continuous releases are steady-state while intermittent releases are non-steady-state and might need to be modelled with non-steady-state models for accuracy in the results.

This review aimed at understanding the potential discrepancies between dispersion modelling using steady-state and non-steady-state models in conditions where meteorological parameters change substantially and gave a qualitative interpretation of the potential differences in impact that can occur. However, a quantification of such discrepancies is necessary before giving thorough recommendations. The development and application of the sensitivity tests to be developed in future work may help to quantify the discrepancies and provide some guidance regarding atmospheric dispersion modelling in changing meteorological conditions.

7 ACKNOWLEDGEMENTS

This review was funded by the Atmospheric Dispersion Liaison Committee (ADMLC). We are grateful to the reviewers for providing useful comments and suggestions on a draft of this report.

8 REFERENCES

Afroz R. M.N Hassan, and N.A Ibrahim, 2003: Review of air pollution and health impacts in Malaysia, Environmental Research, 92, 2, 71-77.

Allen, S.E., 1986: Radiation: a guide to contaminated countryside, The Guardian, 17

Allwine K. J., B.K. Lamb and R. Eskridge, 1992: Wintertime Dispersion in a Mountainous Basin at Roanoke, Virginia: Tracer Study, J. Appl. Meteor., 31, 1295-1311.

Anquetin S, C Guilbaud and J-P Chollet, 1999: Thermal valley inversion impact on the dispersion of a passive pollutant in a complex mountainous area, Atmos. Environ., 33, 3953-3959.

Barad, M. L. (Editor), 1958: Project Prairie Grass, a Filed Program in Diffusion. Geophysical Research Papers, No. 59, Vols I and II. Air Force Cambridge Research Center Report AFCRC-TR-58-235, 479 pages [NTIS PB 151 425 and PB 151 424]

Barclay J., 2008: Low Wind Speed Workshop, MODSIG, CASANZ, Sydney November 2008.

Baumbach G. and U. Vogt, 1999: Experimental determination of the effect of mountain-valley breeze circulation on air pollution in the vicinity of Freiburg, Atmos. Environ., 33, 4019-4027.

BBC, Frost Hollows, http://www.bbc.co.uk/weather/features/understanding/frosthollows.shtml

BBC, Icelandic volcanic ash alert grounds UK flight, 15 April 2010 on http://news.bbc.co.uk/1/hi/8621407.stm

Blewitt D., J. Yohn, R. Koopman, and T.C. Brown, 1987a: "Conduct of Anhydrous Hydrofluoric Acid". International Conference on Vapour Cloud Modelling, Boston, MA, Nov. 2-4, 1987.

Blewitt D., J. Yohn, R. Koopman, T.C. Brown, 1987b: "Effectiveness of Water Sprays on Mitigation of Anhydrous Hydrofluoric Acid Releases', Center for Chemical Process Safety

Bowen B. M., 1996: Example of Reduced Turbulence during Thunderstorm Outflow, J. Appl. Meteor., 35, 1028-1032.

Bowne, N. E., Londergan R.J., Murray D. R., and Borenstein H. S., 1983: Overview, Results and Conclusions for the EPRI Plume Model Validation and Development Project: Plains Site, EPRI EA-3074, Project 1616-1, Elevtric Power Research Institue, Palo Alto, CA. 234pp.

Bozier K., C.G. Collier, F. Davies, A.R. Holt, D.R. Middleton, G.N. Pearson, S. Siement, G. Upton, D.V. Willetts and R.I. Young, 2004: Improved Air Quality Forecasting, Invest to Save Report ISB52-11, Final report for the ISB-52 project (http://www.airquality.co.uk/reports/cat12/0501130859_MS11ISB52ver10.pdf)

Cawthon D., D. Hamlin, A. Steward, C. Davis, F. Cavender, and P. Goad, 2009: Toxicological and Environmental Chemistry. 91, 4.

Cambridge Environmental Research Consultants, Ltd. (CERC), 2010: ADMS 4 User Guide, Version 4.2, February 2010 (http://www.cerc.co.uk/environmental-software/assets/data/doc_userguides/CERC_ADMS4.2_User_Guide.pdf)

Chang Y. S., G. R. Carmichael, H. Kurita and H. Ueda, 1989: The transport and formation of photochemical oxidants in central Japan, Atmos. Environ., 23, Issue, 363-393.

Clappier A., A. Martilli, P. Grossi, P. Thunis, F. Pasi, B. C. Krueger, B. Calpini, G. Graziani and H. van den Bergh, 2000: Effect of Sea Breeze on Air Pollution in the Greater Athens Area. Part I: Numerical Simulations and Field Observations, J. Appl. Meteor., 39, 547-562.

Cook A., P. Willis, C. Witham, 2008: Air Pollution Forecasting: A UK Particulate Episode from 23rd to 24th January 2008 (http://www.airquality.co.uk/reports/cat12/0907301435_Particulate_episode_Jan_08-archive.pdf)

Damato F., Dr.O..Planchon, and V. Dubreuil, 2006: A remote-sensing study of the inland penetration of sea-breeze fronts from the English Channel, Weather, 58, 219-226

Davies F., D.R. Middleton and K.E. Bozier, 2007: Urban air pollution modelling and measurements of boundary layer height, Atmospheric Environment, Volume 41, Issue 19, June 2007, Pages 4040-4049.

Dayal, H., M. Brodwick, R. Morris, T. Baranowski , N.Trieff , J.Harrison, J. Lisse, G. Ansari, 1992, "A Community-based Epidemiologic Study of Health Sequelae of Exposure to Hydrofluoric Acid (HF)", Annal of Epid. 2, 213-230.

Dayan U. and I. Levy, 2005: The Influence of Meteorological Conditions and Atmospheric Circulation Types on PM10 and Visibility in Tel Aviv, J. Appl. Meteor., 44, 606-619.

Dayan U. and D. Lamb, 2007: Influences of atmospheric circulation on the variability of wet sulphate deposition, Int. J. Climatol., DOI: 10.1002/joc.1648.

Dayan U. and D. Lamb, 2005: Global and synoptic-scale weather patterns controlling wet atmospheric deposition over central Europe, Atmospheric Environment, 39, pp521-533.

Draxler R.R., J. L. Heffter, G. D. Rolph, 2002: DATEM – Data Archive of Tracer Experiments and Meteorology, NOAA Air Resources Laboratory, 1315 East West Highway, Silver Spring, Maryland 20910 (http://www.arl.noaa.gov/documents/datem/datem.pdf)

Ferber, G.J., K. Telegadas, J.L. Heffter, C.R. Dickson, R.N. Dietz, P.W. Krey, 1981, Demonstration of a Long-Range Tracer System using Perfluorcarbons, Final Report, January, Tech. Report EPA-600.

Graves H.M., R. Watkings, P. Westbury and P.J., Littlefair, 2001, Cooling Buildings in London, BR431, London, CRC Ltd.

Goldwire H.C. Jr. Et al., 1985, „Desert Tortoise series data report 1983 pressurized ammonia spills", UCID-20562, Lawrence Livermore National Laboratories, Livermore, CA.

Hanna, S.R., D.G. Strimaitis, and J.C. Chang, 1993, "Hazardous Gas Model Evaluation with Field Observations", Atmospheric Environment, Vol. 27A, No.15 pp 2265-2285.

Hanna, S.R., D.G. Strimaitis, and J.C. Chang. 1991, "Evaluation of Fourteen Hazardous Gas Models with Ammonia and Hydrogen Fluoride Field Data" Journal of Hazardous Materials, 26, 127-158

Haugen D.A. (Editor), 1959: Project Prairie Grass, a Field Program in Diffusion. Geophysical Research Papers. No. 59, Vol III. Air Force Cambridge Research Center Report AFCRC-TR-58-235, 673 pages. [NTIS PB 161 101]

Heffter, J.L., J.F. Schubert, and G.A. Meade, 1984, Atlantic Coast Unique Regional Atmospheric Tracer Experiment, October, NOAA Tech Memo. ERL ARL-130.

Hoek M. R., S. Bracebridge, I. Oliver, 2007: Helath impact of the Buncefield oil depot fire, December 2005 - Study of accident and emergency case records. J Public Health, 29 (3), 298-302.

Jones A., Hort M., Manning A., Ryall D., Taylor J., Webster H., Witham C. and Wortley S., 2006: The Buncefield oil depot fire: an overview of actual events and the Met Office's dispersion modelling response. Geophys. Res. Abstracts, 8, 06452.

Kurita H., H. Ueda and S. Mitsumoto, 1990: Combination of Local Wind Systems under Light Gradient Wind Conditions and Its Contribution to the Long-Range Transport of Air Pollutants, J. Appl. Meteor., 29, 331-348

Leach M. J. and A. A. N. Patrinos, 1992: Coastal Circulations and Their Influence on Deposition Patterns in the Washington, D.C. Area, J. Appl. Meteor., 31, 1439-1456

Levy I., U. Dayan and Y. Mahrer (2008), A Five-Year Study of Coastal Recirculation and its Effect on Air Pollutants over the East Mediterranean Region, J. Geophys. Res., 113, D16121, doi:10.1029/2007JD009529.

Lopez M. T., R. Villasenor, A. I. Quintanar, and V. Mora, 2002: Transport and Dispersion of Blowing Dust in the Mexico Basin, Proceeding of ICAR5/GCTE-SEN Join Conference, International Center for Arid and Semiarid Lands Studies, Texas Tech University, Lubbock, Texas, USA Publication 02-2, p330-339.

Luhar A. K. and B. L. Sawford, 1995: Lagrangian Stochastic Modelling of the Coastal Fumigation Phenomenon, J. Appl. Meteor., 34, 2259-2277.

Luhar A. K., P. J. Hurley and K. N. Rayner, 2007: Modelling Low Wind-Speed Stable Conditions in a Prognostic Meteorological Model and Comparison with Field Data, Proceeding of the 11th International Conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes, 2-5 July 2007, 251-255.

Lutman E.R., S. R. Jones, R.A. Hill, P. McDonald, B. Lambers, 2004: Comparison between the predictions of a Gaussian plume model and a Lagrangian particle dispersion model for annual average calculations of long-range dispersion of radionucleides, Journal of Environmental Radioactivity, 75, 339-355.

Maffeis G., Bellasio R. Scire J.S., Longoni M.G., Bianconi R. and Quaranta N., 2001: New Algorithms for the Determination of Solar Radiation, Surface Temperature and PBL Height in CALMET, 25th NATO/CCMS International technical meeting on air pollution modelling and its applications, 15-19 October 2001, Louvain-la-neuve, Belgium

Met Office, the volcano as it develops, 15th April 2010 on http://www.metoffice.gov.uk/corporate/pressoffice/2010/volcano/sat_animation/15Apr/index.html

Moore G. E. and S. D. Reynolds, 1986: Meteorological and air quality analysis in support of the 1985 SCCAMP monitoring program, Documents SYSAPP-86/003, 116pp (Available from Systems Applications Inc, San Rafael, CA 94903).

Muller H. and C. D. Whiteman, 1988: Breakup of a Nocturnal Inversion in the Dischma Valley during DISKUS, J. Appl. Meteor., 27, 188-194.

Murray D.R. and Bowne N.E., 1988: Urban Power Plant Plume Studies, EPRI Report No. EA-5468, Research Project 2736-1, Electric Power Research Institute, Palo Alto, CA.

Nielsen-Gammon, J. W., 2000: The Houston heat pump: modulation of a land-sea breeze by an urban heat island. Preprints, 11th Joint Conf. on the Applications of Air Pollution Meteorology with the A&WMA, Long Beach, CA, Amer. Meteor. Soc., 65-69.

Olesen H. R, P. Loftstrom, R. Berkowicz and M Ketsel, 2005, Regulatory odour model development: Survey of modelling tools and datasets with focus on building effects. , NERI Technical Report No 541 (available on: http://www2.dmu.dk/1_viden/2_Publikationer/3_fagrapporter/rapporter/FR541.pdf)

Orgill, M. M., 1989: Early Morning Ventilation of a Gaseous Tracer from a Mountain Valley, J. Appl. Meteor.,28, 636-651.

Orlanski I., 1975: A Rational Subdivision of Scales for Atmospheric Processes, Bulletin American Meteorological Society, Vol. 56, No. 5, pp527-530.

Robe, F.R., Z-X. Wu and J.S. Scire, 2002: Real-time SO_2 Forecasting System with Combined ETA Analysis and CALPUFF Modelling. Proceedings of the 8th International Conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes, 14-17 October 2002, Sofia, Bulgaria.

Sawford B.L., A.K. Luhar, J.A. Noonan, I.-H. Yoon, S.A. Young, W.L. Physick and G.R. Patterson, J.M. Hacker, J.N. Carras, D.J. Williams, A. Blockley, 1996: Shoreline Fumigation at Kwinana: A Study to assess, validate and improve DISPMOD, Final

Report to Department of Environmental Protection of Western Australia on the Kwinana Fumigation Study, SB/1/227, 323pp

Scire J. S. and J. Chang, 1991: Analysis of Historical Ozone Episodes in the SCCAMP Region and Comparison with SCCAMP 1985 Field Study Data, J. Appl. Meteor., 30, 551-584.

Scire J. S., 2009: Design of a Steady-State index, Conference on Guideline on Air Quality Models: Next Generation of Models, Raleigh, United States, Oct 28-30.

Simpson J. E., 1994: Sea breeze and local winds. Cambridge: Cambridge University Press. XIV + 234 pp.

Smethurst H., C. Witham, A. Robins and V. Murray, 2010: An example of Long-Range Odour Transport, Harmo13, 13th Conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes, Paris, France, 1-4 June, 2010

Smith T. B., 1984: A conceptual model of the air quality environment in Ventura-Santa Barbara Counties. Report Prepared for the Western Oil and Gas Association, 24pp (available from T. B. Smith and Associates, Inc., Pasadena, CA).

Speer M and L. Leslie, 2000: Mesoscale model forecasting as a tool for air pollution management: a case study of sustained smoke pollution over the Greater Sydney area, Meteorol. Appl., 7, 177-186.

Targa J., Kent A., Stewart R., Coleman P., Bower J., Webster H., Taylor J., Murray V., Mohan R. and Aus C., 2006: Initial review of Air Quality aspects of the Buncefield Oil Depot Explosion, AEA/ENV/R/2168, Issue 1, May 2006. (www.airquality.co.uk/archive/reports/cat05/0606201126_Buncefield_report_vF3_text2.pdf)

Tinarelli G., Anfossi D., Brusasca G., Ferrero E., Giostra U., Morselli M.G., Moussafir J., Tampieri F., Trombetti F., 1994: Lagrangian particle simulation of tracer dispersion in the lee of a schematic two-dimensional hill, Journal of Applied Meteorology, 33, N.6, 744-756.

Tinarelli G., D. Anfossi, M. Bider, E. Ferrero, S. Trini Castelli, 2000: A new high performance version of the Lagrangian particle dispersion model SPRAY, some case studies. Air Pollution Modelling and its Applications XIII, S.E. Gryning and E. Batchvarova eds., Kluwer Academic / Plenum Press, New York, 499-507.

Triantafyllou A. G., E. S. Kiros, and V. G. Evagelopoulos, 2002: Respirable Particulate Matter at an Urban and Nearby Industrial Location: Concentrations and Variability and Synoptic Weather Conditions during High Pollution Episodes, J. Air & Waste Management Association, 52: 174-185

U.S. Environmental Protection Agency (USEPA), 2003: AERMOD: Latest Features and Evaluation Results, Technical document EPA-454/R-03-003, June 2003 (http://www.epa.gov/scramm001/7thconf/aermop_mep.pdf

Vautard R., P. Ciais, R. Fisher, D. Lowry, F.M. Breon, F. Vogel, I Levin, F. Miglietta and E. Nisbet, 2007: The Dispersion of the Buncefield oil fire plume: An extreme accident without air quality consequences, Atmos. Environ, 41, 9506-9517.

Venkatram A., 2002, Accounting for averaging time in air pollution modelling, Atmospheric Environment, 36, 2165-2170

WGN Weather, 2008, Lake breeze and urban 'heat island' enhance downpours over Chicago, http://weblogs.wgntv.com/chicago-weather/tom-skilling-blog/2008/08/lake-breeze-and-urban-heat-isl.html

Whiteman C. D., 1989, Morning Transition Tracer Experiments in a Deep Narrow Valley, J. Appl. Meteor., 28, 626-635.

Witham C., 2008: The influence of long-range transport in recent UK air pollution events, Air Quality Forecasting Seminar, 14 May 2008

(http://www.airquality.co.uk/reports/cat12/0805191156_8_Witham_LongRangeAQTransport.pdf)

Wilby L., 2003, Past and projected trends in London's urban heat island, Weather, 58

Zhang J. and S. T. Rao, 1999: The Role of Vertical Mixing in the Temporal Evolution of Ground-Level Ozone Concentrations, J. Appl. Meteor., 38, 1674-1691.

9 TABLES

Table 1 Meteorological conditions and applications classified by Time Scale

Time Scale	Change in Meteorological conditions	Applications
Minutes to one hour	- Gust front / outflow boundary - Thunderstorm / Squall lines - Light wind speed – meandering - Change in Precipitation	- Short-term accidental release - Odour modelling - forecast modelling - real-time operational modelling - Long-range modelling (local changes along the path after release)
Hours to one day	- Air Recirculation (Land/Sea breeze; Mountain/Valley flow,...) - Urban Heat Island effect - Inversion break-up fumigation - Shoreline fumigation - Light wind speed follow by frontal gust - Change in Precipitation	- Short-term accidental release - Odour Modelling - Forecast modelling - Real-time operational modelling - Regulatory Impact Assessment (hourly averaged impact) - Cumulative Impact Assessment - Long-range modelling (local changes along the path after release) - Risk Assessment - Volcanic Eruption, Fire
A few days	- Recirculation happening on a number of consecutive days (Land/Sea breeze, Mountain/Valley flow,...) - Anticyclone situation and the sub-meteorological conditions that can happen under this situation (radiation temperature inversion in winter - Succession of frontal passages	- Regulatory Impact assessment (daily averaged impact) - Cumulative Impact Assessment - Risk Assessment - Long-range accidental release - Volcanic Eruption, Fire
Months/Annual	- High frequency of any of the above	- Regulatory Impact Assessment (monthly or seasonal averages, annual averages) - Cumulative Impact Assessment - Risk Assessment - Long-range Accidental release - Volcanic Eruption, Fire

Table 2 Changing meteorological conditions and their possible impact on receptors and type of applications

Meteorological Parameter	Change in Meteorological conditions	Consequences on Receptors	Applications
Change in Wind Direction	- Gust front / Outflow boundary - Thunderstorm / Squall lines - Urban Heat Island effect	- Change the location of impact	- Short-term accidental release - Odour modelling - forecast modelling - real-time operational modelling - Long-range applications (local changes along the path after release) - Volcanic Eruption, Fire, Sand Transport - Cumulative Impact Assessment
	- Light wind speed – meandering - Air Recirculation (Land/Sea Breeze; Mountain Valley flow, etc…) - Light wind speed follow by frontal gust	- Create pollutant accumulation and so subject to potential high concentrations	
Change in Wind Speed	- Gust front / outflow boundary - Thunderstorm / Squall lines - Air Recirculation (Land/Sea breeze; Mountain/Valley flow, etc…) - Urban Heat Island effect - Light wind speed follow by frontal gust	- Change in dilution - Change in dispersion - Change in distance from the source maximal impact	- Short-term accidental release - Odour Modelling - Regulatory Impact Assessment - Forecast modelling - Real-time operational modelling - Regulatory Impact Assessment (hourly averaged impact) - Cumulative Impact Assessment - Long-range modelling (local changes along the path after release) - Risk Assessment - Volcanic Eruption, Fire, Sand Transport
Change in Mixing Height Or Change in Turbulence / Stability class	- Urban heat Island effect - Inversion break-up fumigation - Shoreline fumigation - Land/Sea Breeze	- Change in ground concentration - Change in dilution - Change in dispersion	- Elevated sources - Regulatory Impact assessment (daily averaged impact) - Cumulative Impact Assessment - Risk Assessment
Change in Precipitation	- Thunderstorm / Squall lines - Frontal Passage	- Wet deposition - Remove pollutant material from air along path	- Volcanic Eruption, Fire, Sand Transport - Long-range applications (local changes along the path after release) - Regulatory Impact Assessment (hourly /daily / annual averaged impact)

Table 3 Proposed studies for testing steady-state versus non-steady-state models

Test	Dataset	Change in Meteorological conditions	Time Scale	Distance from Source	Impact	Comments
Test 1	Kwinana	Shoreline Fumigation	Short-Term (1h-, 24h-averages)	Near-field (within 10km)	Peak Concentration	Coastal location, under sea-breeze conditions
Test 2	Tracy Power Plant	Morning Fumigation	Short-Term (1h-average)	Near-field (within 10km)	Peak Concentration	Complex Terrain study – not exactly strictly variation due to meteorology
Test 3	ISB52	Passage of Front / Precipitation	Short-Term (1h-, 24h-averages)	Near-field (within 10km)	Path / Wet Deposition	Only meteorological data available
Test 4	Cardington, UK	Low wind speed conditions	Short-Term (sub-hourly, 1h-average)	Near-field (within 10km)	Peak Concentration	Only meteorological data available
Test 5	ISB52	Morning Fumigation	Short-Term (1h-average)	Near-field (within 10km)	Peak concentration / Path	Only meteorological data available

10 FIGURES

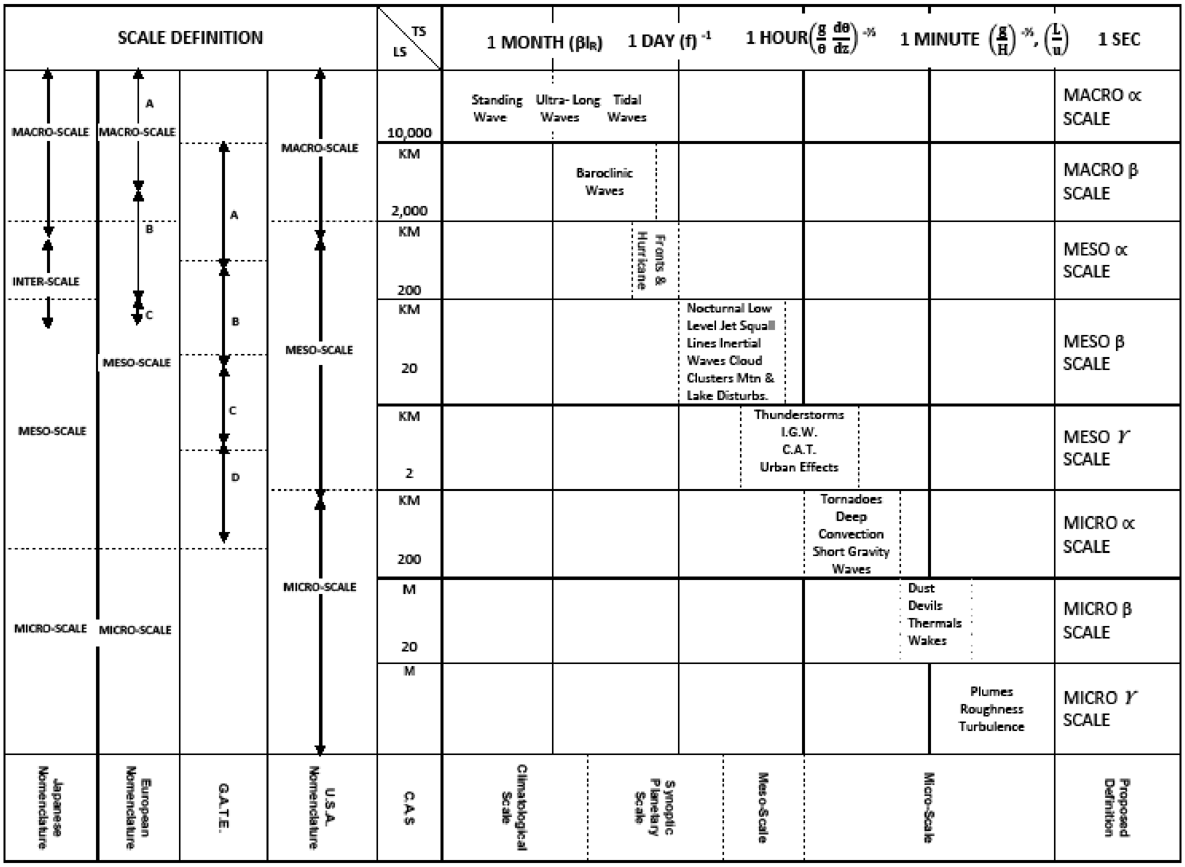


Figure 1 Atmospheric Dynamic Spectrum classified by temporal and spatial scales (reproduced from Orlanski, 1975) – The terms in parenthesis along the timescale row are physical parameters known to be controlling each particular range of time scales.

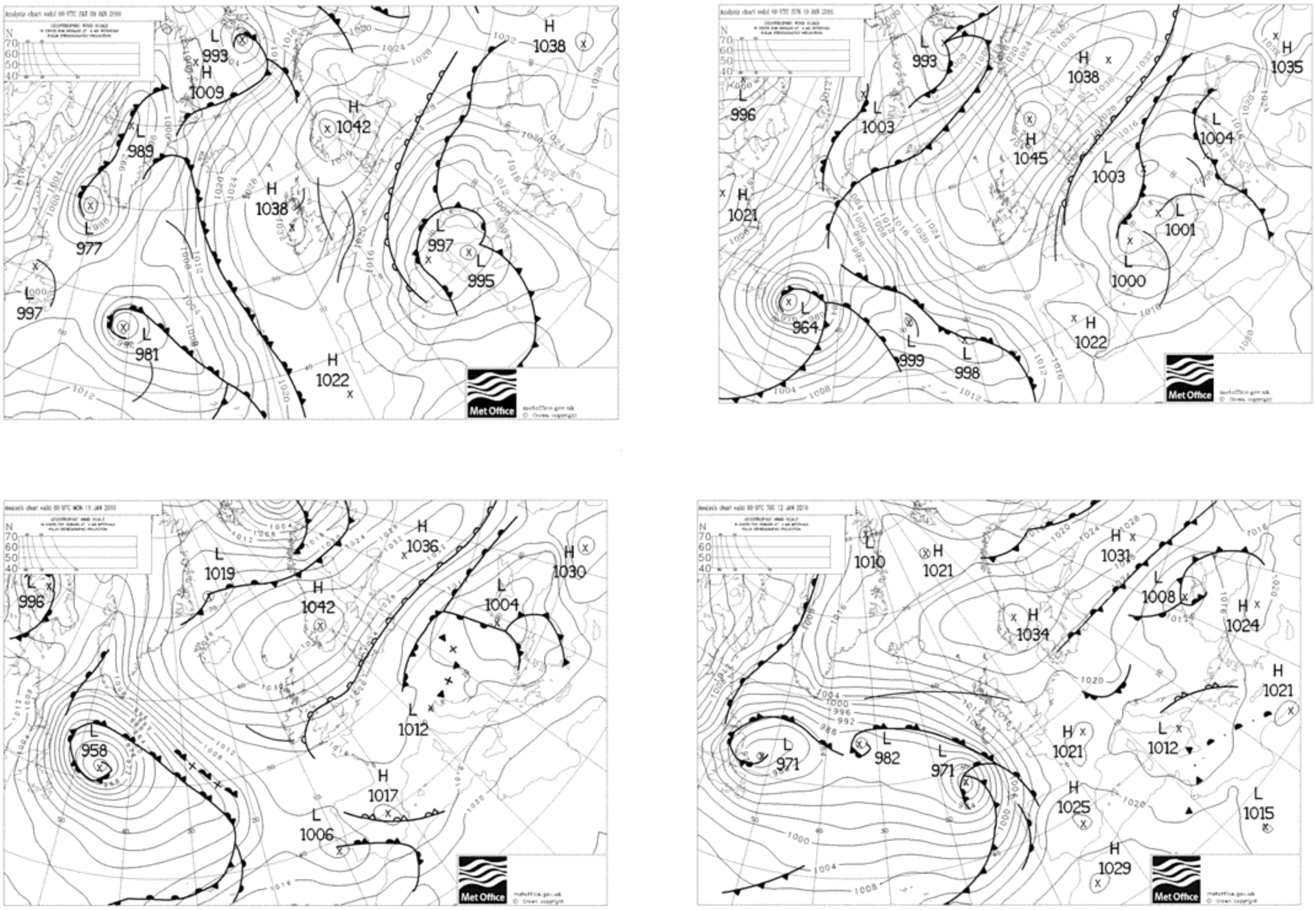


Figure 2 Example of stationary High Pressure centred on Norway for a 4-day period (January 9, 2010 (top left), January 10, 2010 (top right), January 11, 2010 (bottom left) and January 12, 2010 (bottom right)

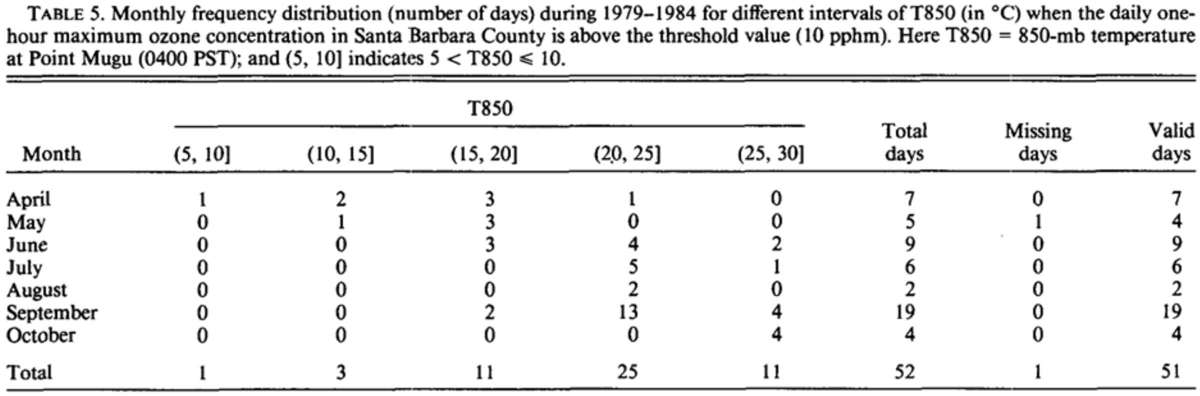


TABLE 5. Monthly frequency distribution (number of days) during 1979–1984 for different intervals of T850 (in °C) when the daily one-hour maximum ozone concentration in Santa Barbara County is above the threshold value (10 pphm). Here T850 = 850-mb temperature at Point Mugu (0400 PST); and (5, 10] indicates 5 < T850 ≤ 10.

	T850							
Month	(5, 10]	(10, 15]	(15, 20]	(20, 25]	(25, 30]	Total days	Missing days	Valid days
April	1	2	3	1	0	7	0	7
May	0	1	3	0	0	5	1	4
June	0	0	3	4	2	9	0	9
July	0	0	0	5	1	6	0	6
August	0	0	0	2	0	2	0	2
September	0	0	2	13	4	19	0	19
October	0	0	0	0	4	4	0	4
Total	1	3	11	25	11	52	1	51

Figure 3 Table 5 (From Scire and Chang, 1991) shows how high ozone concentrations happen when the temperature at 850mb is usually high. The maximum occurrences are recorded in the month of September

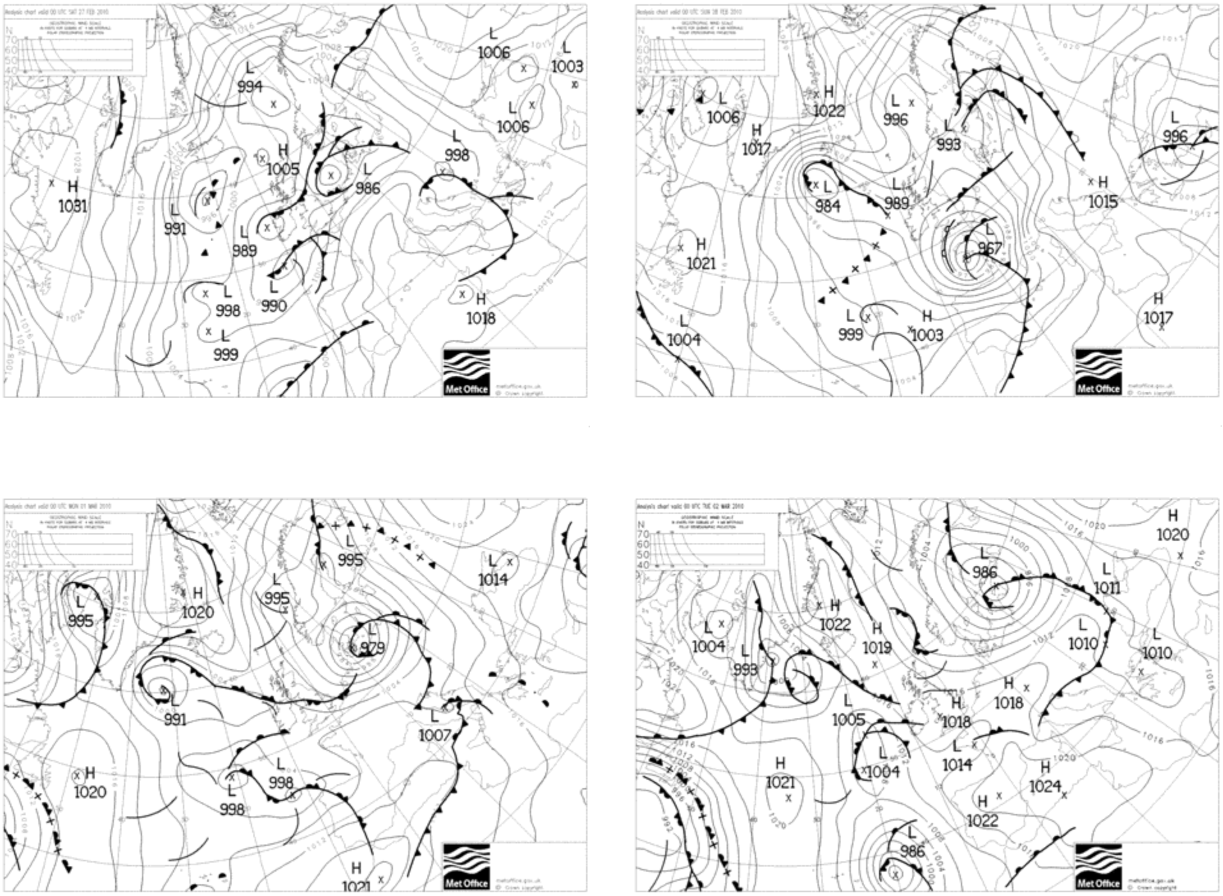


Figure 4 Example of a fast moving Low pressure from the South West toward the North East region of Europe (Feb 27, Feb 28 and March 1, March 2, 2010)

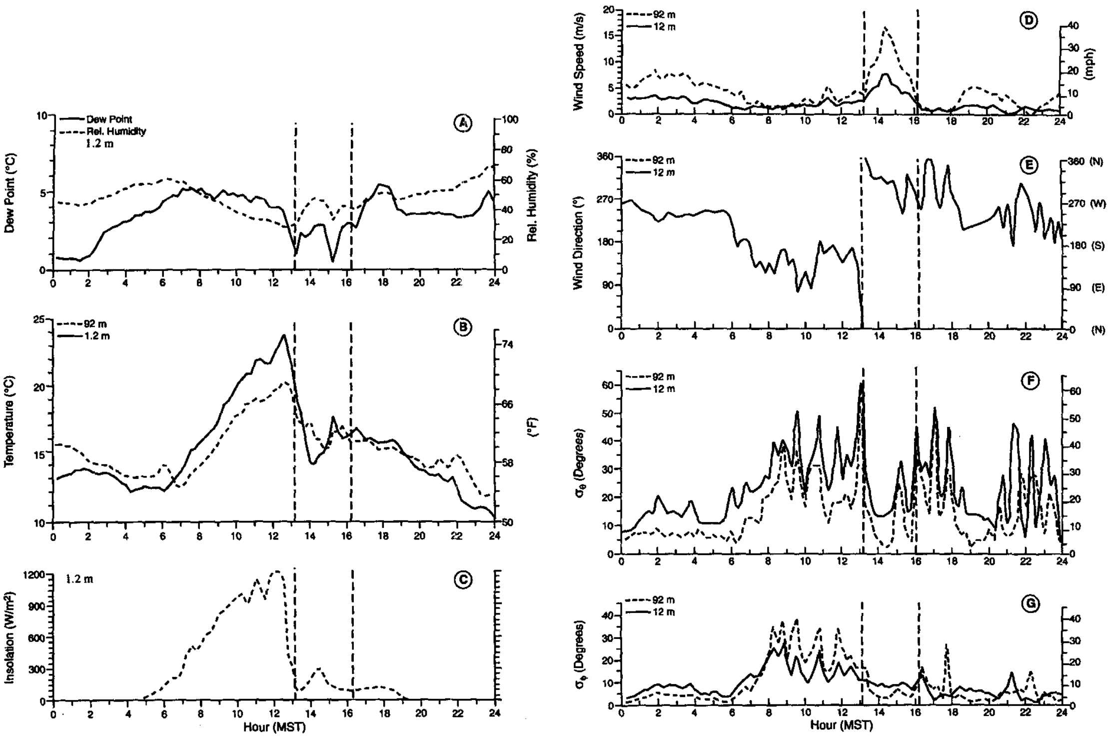


Fig. 2. Tower measurements during thunderstorm flow on 9 June 1991. Analyses include 15-min averages of (a) dewpoint and relative humidity, (b) temperature, (c) insolation, (d) wind speed, (e) wind direction, (f) standard deviation of wind direction (σ_θ), and (g) standard deviation of vertical wind direction (σ_ϕ). Measurement levels above ground are shown in upper-left corners of each plot. Vertical dotted lines indicate estimated onset and cessation times of outflow.

Figure 5 Plots showing the passage of an outflow of a weak thunderstorm on Jun 9, 1991 between approximately 1300 and 1630 local time (From Bowen, 1996)

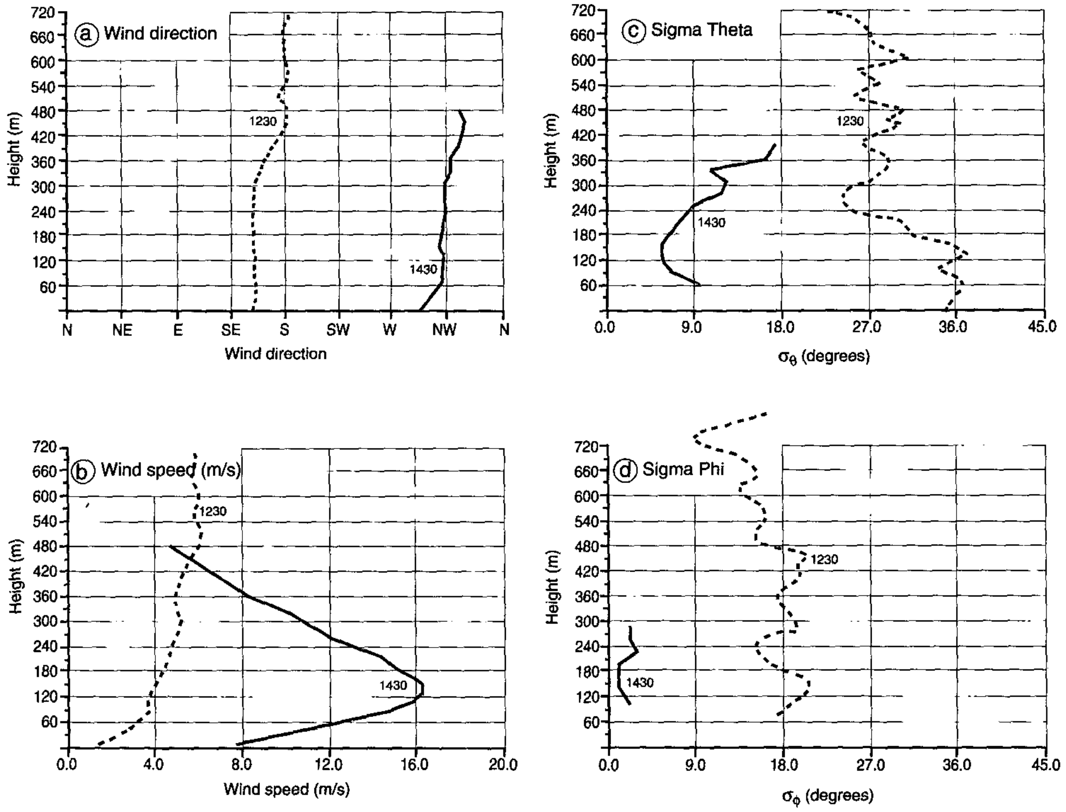


FIG. 3. Vertical profiles of (a) wind direction, (b) wind speed, (c) σ_θ, and (d) σ_ϕ measured by sodar before (1230 LST) and after (1430 LST) gust front passage on 9 June 1991.

Figure 6 Four plots showing vertical profile of wind speed, wind direction and horizontal and vertical variation of wind before (1230 LST) and during (1430 LST) the passage of an outflow of a weak thunderstorm on Jun 9, 1991 (From Bowen, 1996)

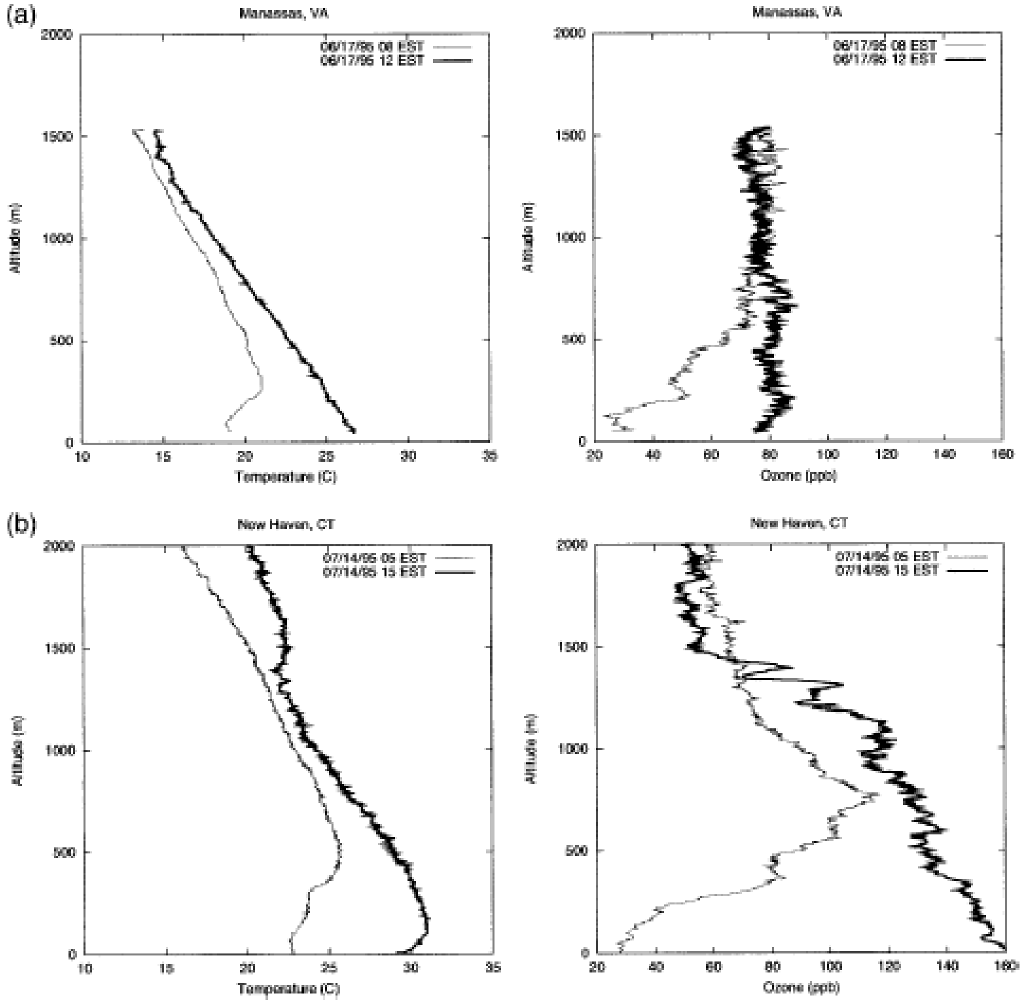


FIG. 8. Aircraft measurements of temperature and ozone profiles at (a) Manassas, Virginia, on 17 June 1995 and (b) New Haven, Connecticut, on 14 July 1995.

Figure 7 Figure 8 from Zhang and Rao (1999) shows the correlation of change in temperature vertical profile and ozone vertical profile for two separate events (17 June 1995 at Manassas, VA (top) and 14 July 1995 at New Haven, Connecticut (bottom)).

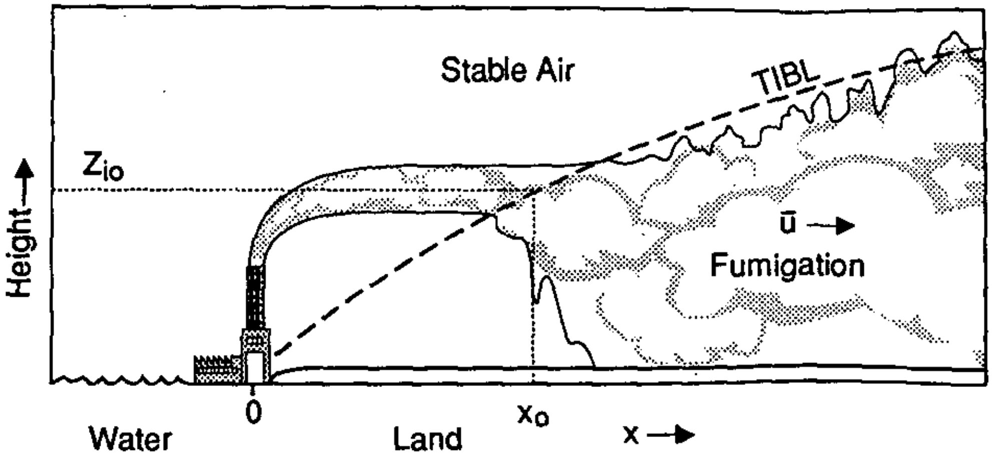


FIG. 1. Illustration of the coastal fumigation phenomenon. The TIBL is shown by a dashed line.

Figure 8 **Schematic illustration of coastal fumigation (from Luhar and Sawford, 1995)**

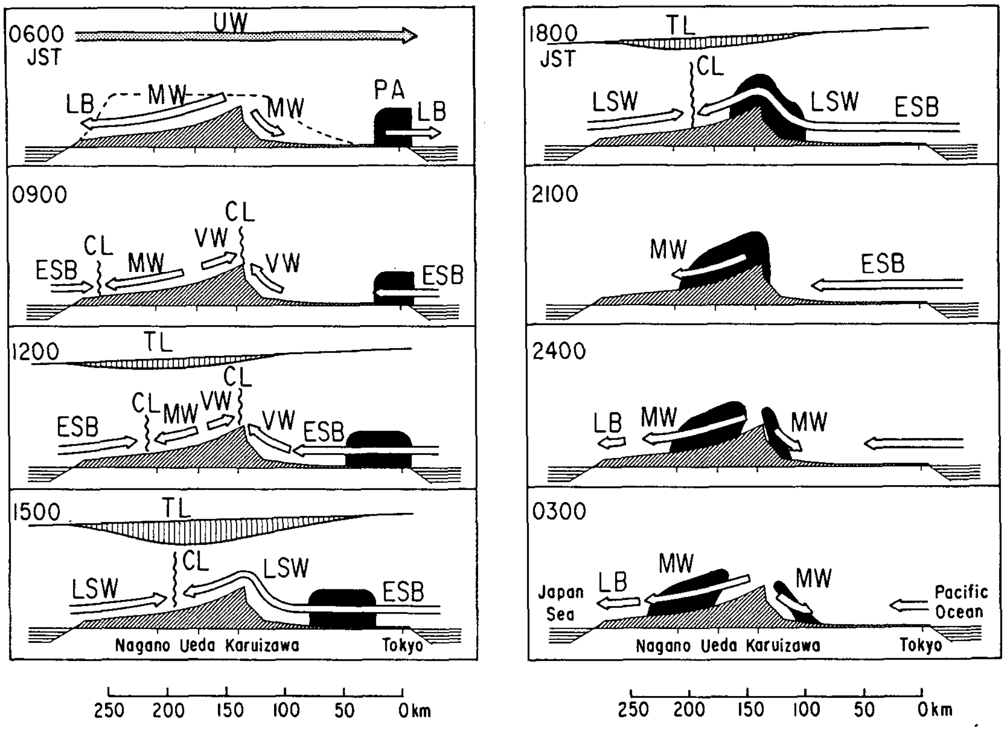


FIG. 16. Schematic diagram of the transport process of the air pollutants from the coastal region to the mountainous inland region. Broken line denotes the average altitude of the mountainous central region. Vertical hatching: a depression in sea level pressure; PA: polluted air; TL: thermal low; CL: convergence line; UW: upper wind (only shown for 0600 JST); LB: land breeze; MW: mountain wind; VW: valley wind; ESB: extended sea breeze; LSW: large-scale wind toward the thermal low.

Figure 9 Figure 16 from Kurita et al., 1990 shows the diurnal variation of the combination of meteorological events resulting in diurnal changing meteorological conditions.

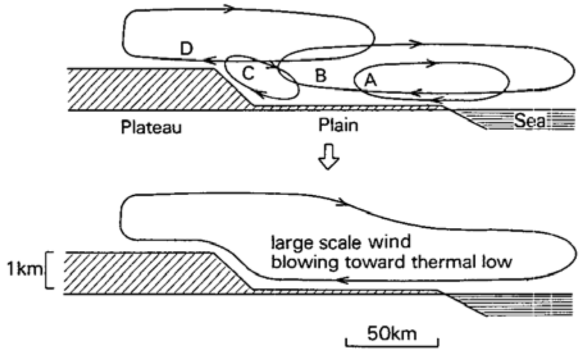


FIG. 15. Schematic diagram of the combination of the local wind systems and the generation of the large-scale wind system drawn toward the thermal low. A, B, C and D correspond to the letters in Table 1.

TABLE 1. Classification of the local and large-scale wind systems. *A*: land/sea breeze circulation caused by the temperature difference between land and sea (diurnal temperature variation). *B*: onshore wind caused by the diurnal-mean temperature difference between land and sea (Ueda 1983). *C*: slope/valley wind caused by heating of the mountain slope. *D*: plain/plateau wind circulation caused by the temperature difference between plain and plateau (Mannouji 1982; Arisawa 1987; Ueda et al. 1988b).

Phenomenological classification	Composition
Sea breeze	A
Extended sea breeze	$B + A$
Valley wind (upslope wind)	C
Large-scale wind blowing into thermal low	$D + C\ (+B + A)$

Figure 10 Figure 15 from Kurita et al., 1990 shows a combination of meteorological events (land / sea breeze, onshore wind, slope / valley wind and plain / plateau wind) resulting in changing meteorological conditions.

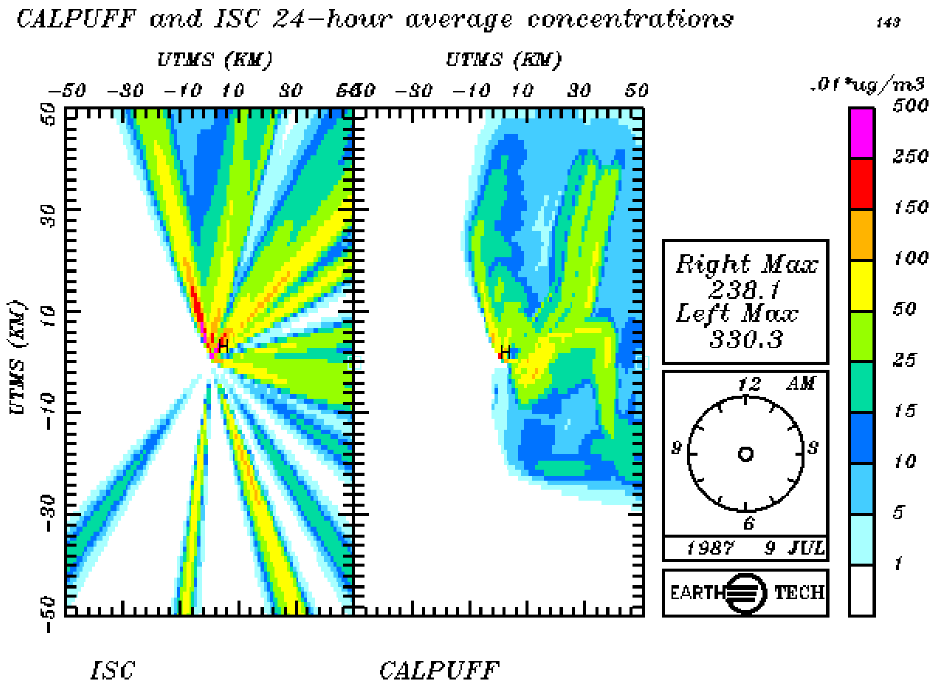


Figure 11 24h-average SO_2 concentration simulated using a simple Gaussian model (steady-state) on the left and a Lagrangian puff model (non-steady-state) on the right (from animation developed for CALPUFF Training by the Atmospheric Study Group (ASG), Earth Tech.). This Figure shows the potential 24h concentration footprint discrepancies between a steady-state model and a non-steady-state model

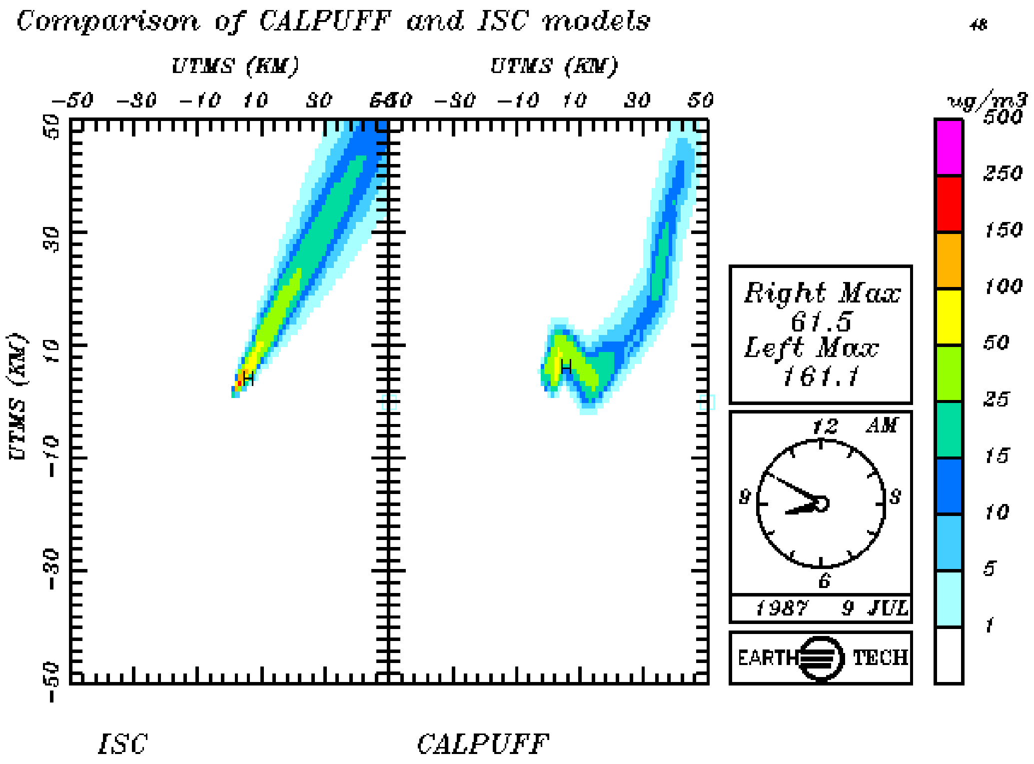


Figure 12 hour 9 average SO_2 concentration simulated using a simple Gaussian model (steady-state) on the left and a Lagrangian puff model (non-steady-state) on the right (from animation developed for CALPUFF Training by the Atmospheric Study Group (ASG), Earth Tech.). This figure shows much larger concentration for the steady-state model and a curved trajectory for the non-steady-state model

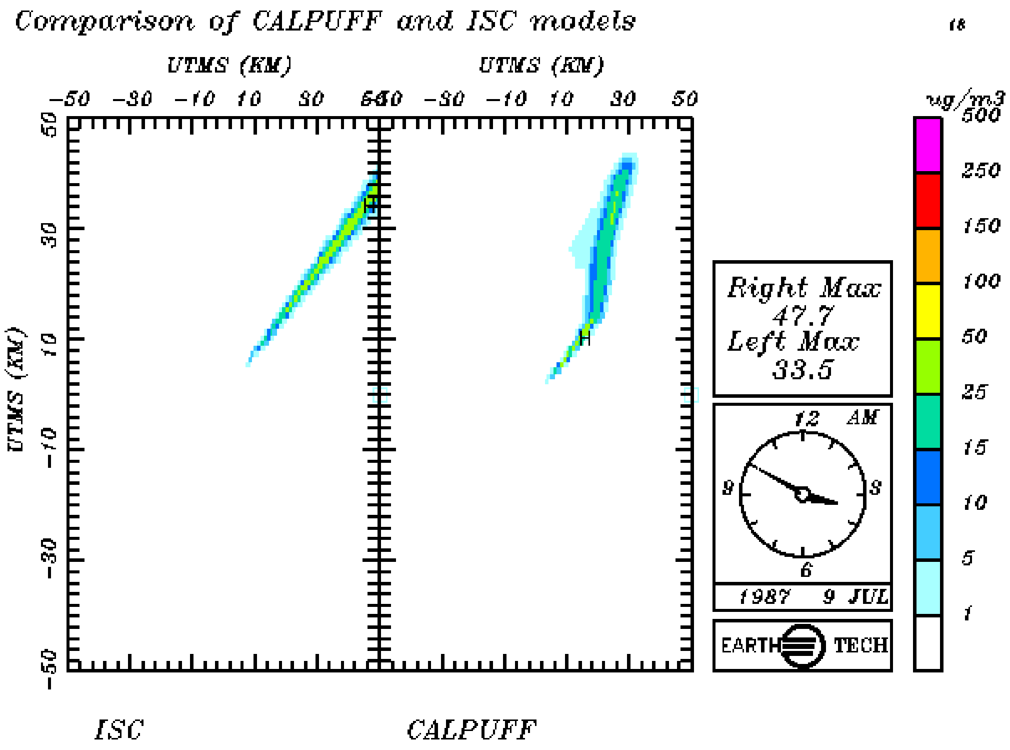


Figure 13 Hour 4 average SO_2 concentration (a few hours earlier than Figure 12) simulated using ISC, a simple Gaussian model (steady-state) on the left and CALPUFF, a Lagrangian puff model (non-steady-state) on the right (from animation developed for CALPUFF Training by the Atmospheric Study Group (ASG), Earth Tech.). This plot shows lower concentrations for the steady-state model and completely different location of impact. On the right side, there has been accumulation of concentration. On the left side, the highest impact is at the edge of the domain.

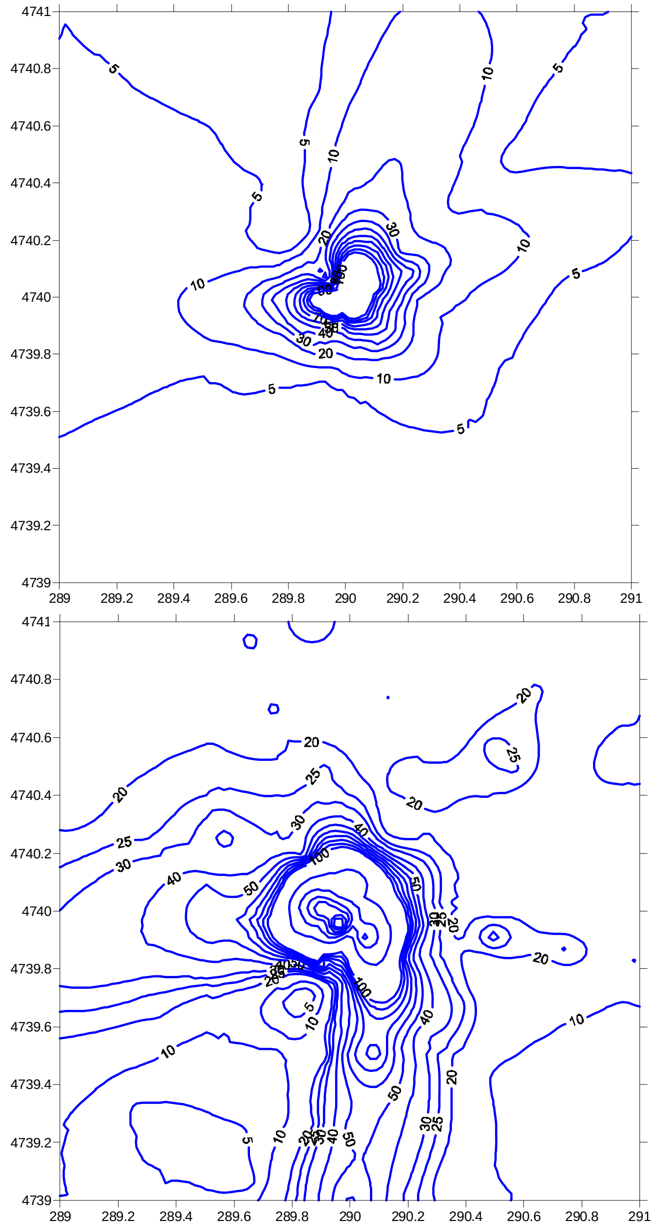
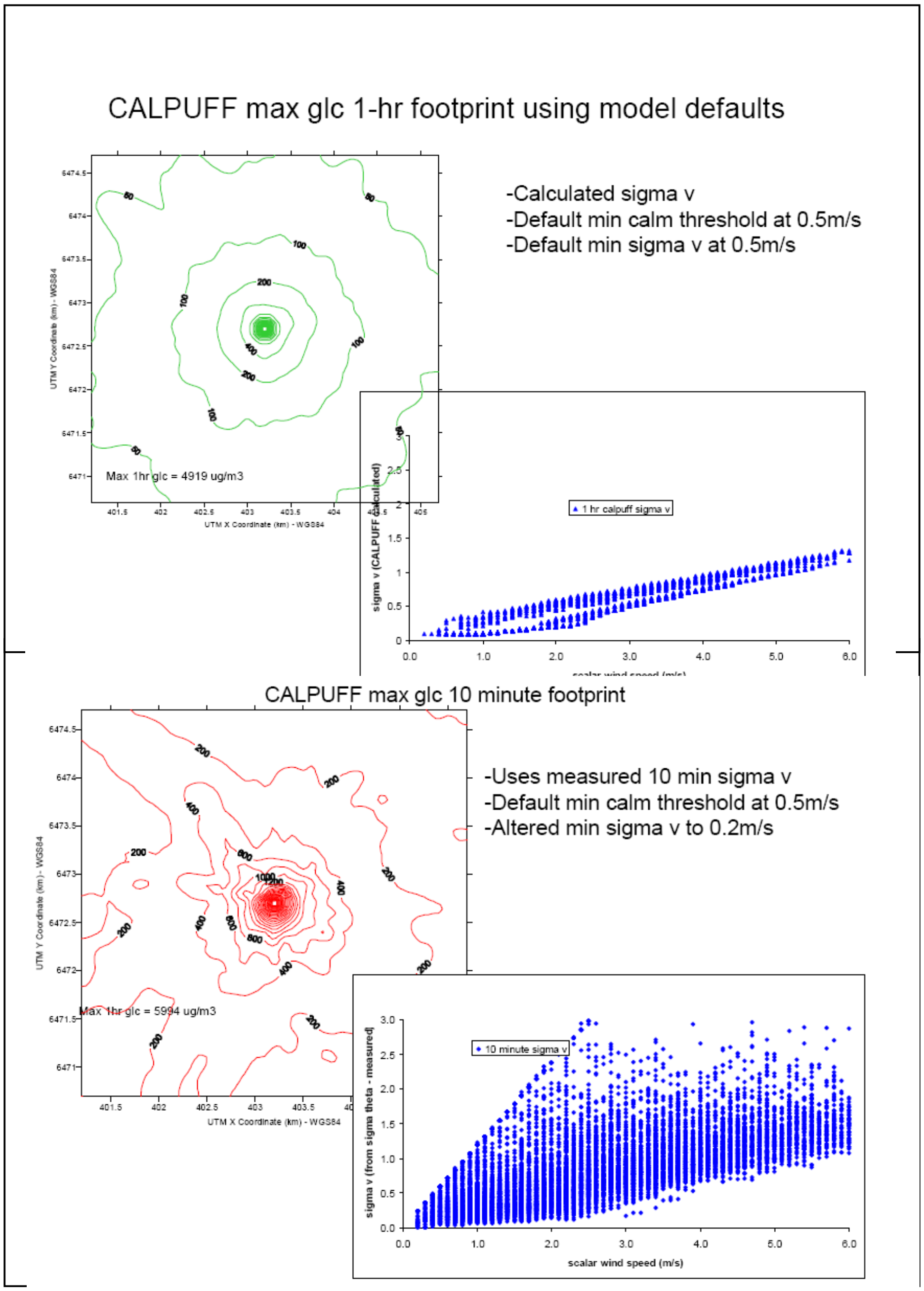


Figure 14 H$_2$S averaged concentration on the period 8/8 (16h) to 8/10 (10h), year 2006, modelled by CALPUFF using model defaults - hourly meteorological data, horizontal sigma=0.5 m/s and a calm threshold of 0.5m/s (top), versus 6-minutes average meteorological data, horizontal sigma=0.2 m/s and a calm threshold of 0.5m/s (bottom) (From Barclay, 2008).



CALPUFF max glc 1-hr footprint using model defaults

-Calculated sigma v
-Default min calm threshold at 0.5m/s
-Default min sigma v at 0.5m/s

CALPUFF max glc 10 minute footprint

-Uses measured 10 min sigma v
-Default min calm threshold at 0.5m/s
-Altered min sigma v to 0.2m/s

Figure 15 1-hour peak ground level H_2S concentration modelled using model defaults - hourly meteorological data, a calm wind speed threshold of 0.5 m/s, minimum horizontal sigma = 0.5 m/s and internally computed turbulence parameters (top) versus 10-min peak ground level H2S concentration using 10 minute meteorological data, a calm wind speed threshold of 0.5 m/s, minimum horizontal sigma = 0.2 m/s and real time turbulence parameters (bottom) – Both are computed with CALPUFF code on the year 2006 period: 8/8 at 16h to 8/10 at 10h (From Barclay, 2008).

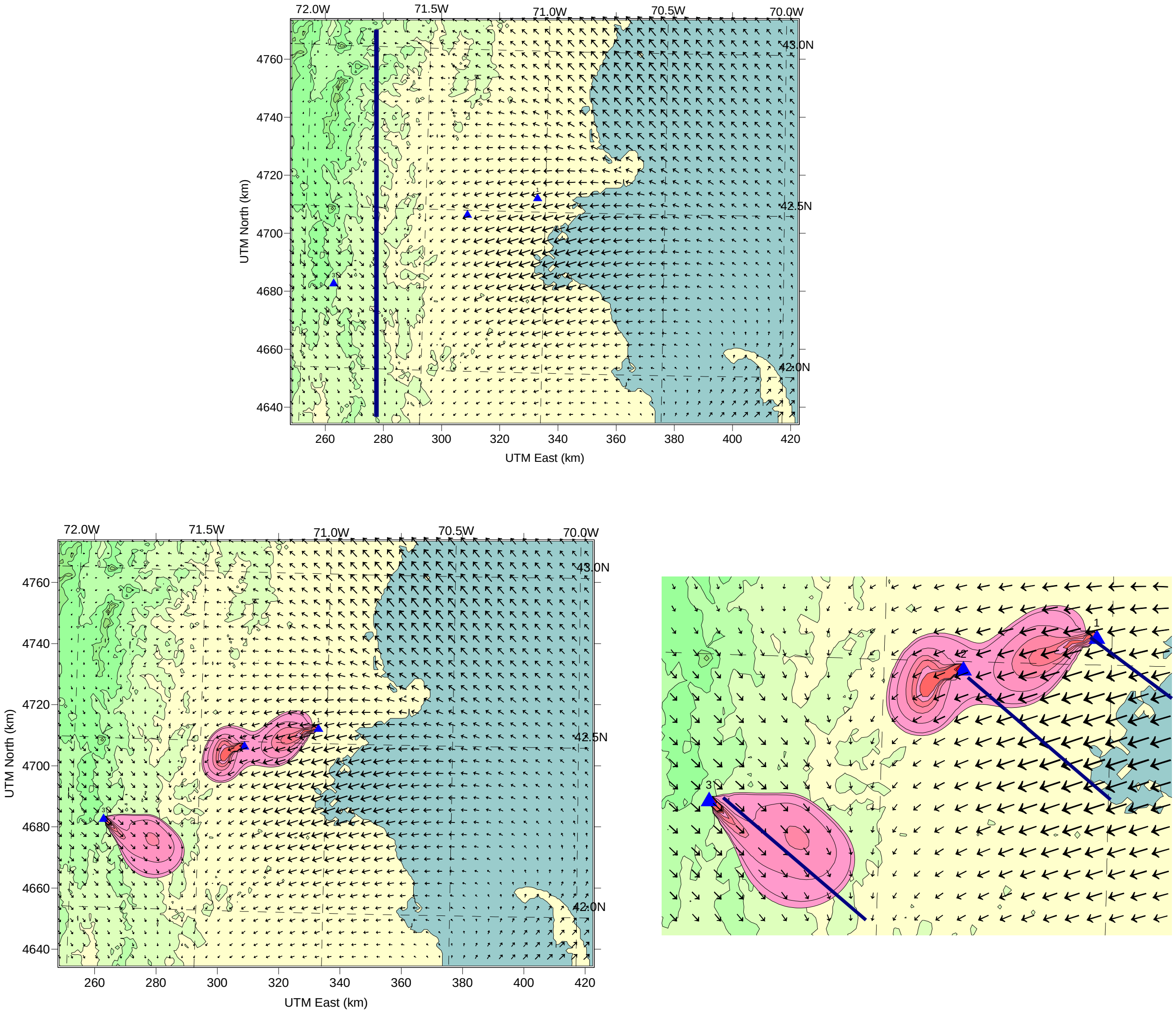


Figure 16 Cumulative Impact Assessment for NAAQS compliance – The facility of interest is on the Western side, embedded in a land breeze circulation (July 7, 1988 – 1pm Local Time). Two neighbouring facilities are located closer to the coast and experience sea breeze conditions. Straight plume model AERMOD's trajectories (shown as blue lines on the bottom right picture), using meteorological information from the most inland station, do not reflect the current situation; three-dimensional Lagrangian puff model CALPUFF's impacts do (pink ground concentration contours) - (From Scire, 2009)

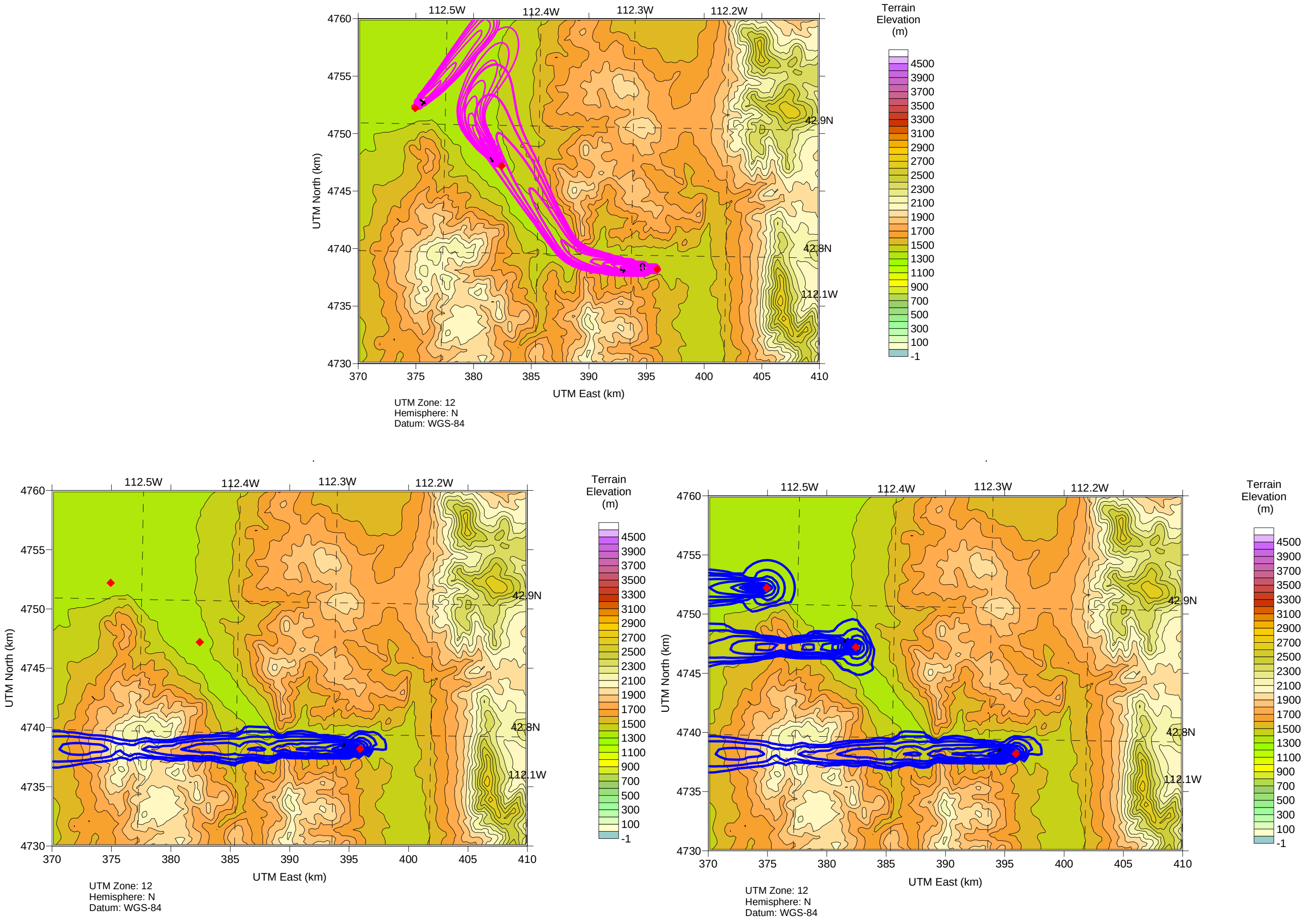


Figure 17 Terrain Channelling Effect – The facility of interest (INKOM) is located within a deep curving valley. Straight plume modelling with AERMOD (bottom left, blue) shoots the INKOM plume toward the valley sides and over the mountains. Lagrangian puff modelling with CALPUFF (top, pink) correctly models the curved trajectories. The Straight plume model (AERMOD) also fails to correctly model the cumulative impact of the other 2 sources in the area, FMC, HOSP (bottom right) – (From Scire, 2009)

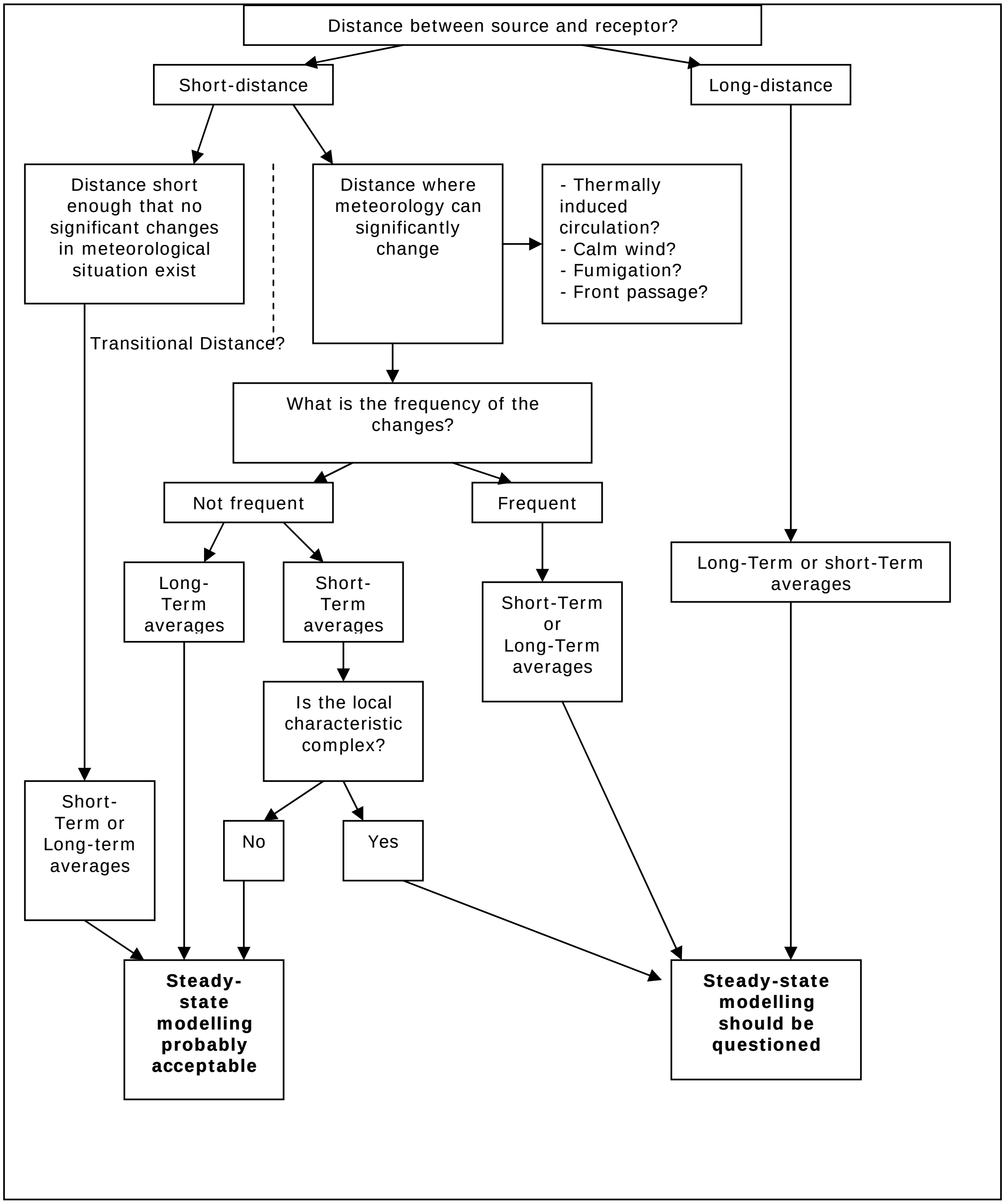


Figure 18 **Proposed procedure to determine whether a steady-state model or a non-steady-state model should be used for the application of concern.**