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The use of long-range transport simulations to verify the Australian Air Quality Forecasting System

K.J. Tory
Bureau of Meteorology Research Centre, Australia

M.E. Cope
CSIRO Atmospheric Research, Australia and CSIRO Energy Technology, Australia

G.D. Hess
Bureau of Meteorology Research Centre, Australia

S. Lee
CSIRO Atmospheric Research, Australia
and

N. Wong
Environment Protection Authority of Victoria, Australia

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The Australian Air Quality Forecasting System (AAQFS) was used to model long-range transport of carbon monoxide (CO) from Melbourne to Cape Grim, a distance of about 300 km. Three periods of elevated CO concentrations were observed at Cape Grim during a 13-day study period that ran from 18 September to 1 October, 2001. The AAQFS successfully predicted elevated CO concentrations during all three periods when a detecting radius of 0.1° (about 10 km) was used. This is a very good result given the expected trajectory error over that distance is about 100 km. A more quantitative assessment revealed timing errors of up to 12 hours, and the modelled peaks in the Cape Grim CO time-series tended to be shorter in duration and greater in magnitude when compared with that observed. The timing errors were attributed to errors in forecasting the wind trajectories.

It is suggested that the forecasts could be improved by increasing the horizontal diffusion to reduce the 'peakiness' of the modelled CO time-series, and by reducing a slight overestimation of the height of the marine boundary layer (to increase modelled concentrations by reducing the vertical dilution).

Although unverifiable in detail due to insufficient observations, the AAQFS predicted a number of interesting plume features. These include pulsing of CO out of the Melbourne airshed due to morning and evening peaks in CO production that propagated across Bass Strait as distinct plumes, and plume folding and sweeping back and forth across Bass Strait due to spatial and temporal variations in wind direction.

Introduction

The Australian Air Quality Forecasting System (AAQFS) is a joint project between the Bureau of Meteorology, CSIRO Atmospheric Research, CSIRO Energy Technology, the Environment Protection Authority Victoria and the New South Wales Environment Protection Authority to develop a high-resolution numerical air quality forecasting system. Currently 24 to 36-hour forecasts are produced in both Melbourne and Sydney twice daily at a horizontal resolution of 1 km in urban areas and 5 km in rural areas. Forecasts are provided for a number of species including, oxides of nitrogen (NO_x), ozone (O_3), sul-

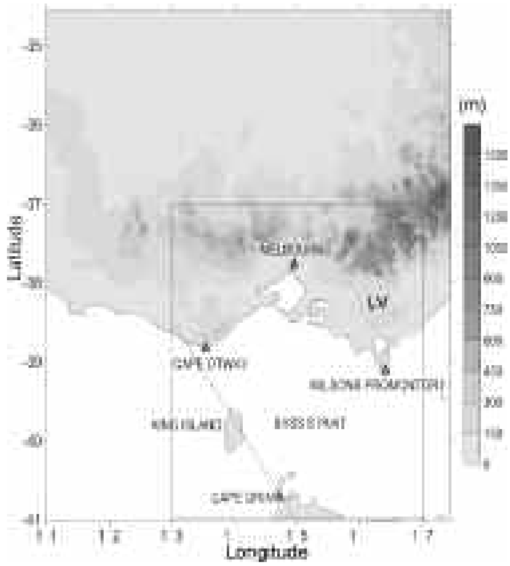
Corresponding author address: K.J. Tory, Bureau of Meteorology Research Centre, GPO Box 1289K, Melbourne, Vic. 3001, Australia. Email: k.tory@bom.gov.au

phur dioxide (SO_2), carbon monoxide (CO), benzene (C_6H_6), formaldehyde (CH_2O), inhalable particles with diameters < 10 μm (PM10), respirable particles with diameters < 2.5 μm (PM2.5) and passive tracers if required. A description of the complete system and its performance is given by Manins (2001).

Long-range pollutant transport is a problem of particular concern to air quality (AQ) forecasters in many parts of the world (in this paper we use the term 'long-range' to represent distances of 200 km and above). Regions far downstream can be affected by particularly large sources, such as are found in North American, European, Chinese and Japanese cities (e.g. Kallos et al. 1998; Jaffe et al. 1999; Fenneteaux et al. 1999; Wanger et al. 2000; Kato et al. 2001). In these cases the AQ forecast-model domains need to be large enough to include all sources that are likely to influence the forecast location, or include some form of parametrisation of external sources at the model boundaries. In Australia, AQ forecasting in the major population centres is largely free of such problems. The large distances between these centres in Australia and the relative isolation of Australian cities from pollutant sources outside Australia, have enabled the development of a numerical forecasting system on domains that include all significant pollutant sources, and still remain small enough to be run twice per day.

Recognising the importance of long-range transport in other parts of the world, we believe it is useful to investigate the ability of the AAQFS to forecast long-range transport. A valuable opportunity exists to test long-range transport of various gaseous pollutants and particles between Melbourne and Cape Grim (Fig. 1). Cape Grim is located at the northwest tip of Tasmania about 320 km south of Melbourne, and it houses the Cape Grim Baseline Air Pollution Station (CGBAPS). CGBAPS monitors global atmospheric composition and meteorological conditions. In this paper we investigate the urban plume transport of CO from the Melbourne airshed to Cape Grim. In the Melbourne airshed CO is primarily generated by motor vehicles and is quasi-conservative over timescales representative of this study. The path between the Melbourne airshed and Cape Grim is almost entirely over sea surface (Bass Strait), and is (nearly) free of other CO sources. Additional sources of CO come from Victorian regional centres and the Latrobe Valley where brown coal is burned to generate the majority of Melbourne's electrical power. The Latrobe Valley is located about 150 km to the east-southeast of Melbourne (Fig. 1). In this study the forecast CO contribution from the Latrobe Valley is insignificant compared with Melbourne, and a visual examination of Figs 3 to 6 (introduced later) shows no significant CO transport from this location to Cape Grim. Contributions from the regional centres are even smaller than those from the Latrobe Valley. Hereafter, the name Melbourne when used in the context of airsheds or emission sources includes Melbourne, Latrobe Valley and other regional emissions. The emissions inventory of Ng et al. (2000) provides the Melbourne CO sources used in this work.

Fig. 1 Grid 1 topography. The rectangular box shows the subsection of Grid 1 used in Figs 3 to 5. The diagonal line that intersects King Island and Cape Grim marks the location of the vertical section presented in Fig. 6. The Latrobe Valley location is indicated by 'LV'.



Cox et al. (1999) also investigated the Melbourne to Cape Grim CO transport in their model TAPM (a different meteorological and dispersion model driven by meteorological analyses, rather than forecasts). Their study ran for 18 months (March 1998 to September 1999) and focussed more on the climatology of CO pollution at Cape Grim. They found that over 80 per cent of the major CO pollution episodes observed at Cape Grim were influenced by the Melbourne airshed. In the current study a 13-day period, beginning on 18 September 2001, is investigated in which three distinct periods of elevated CO concentrations are present in the CGBAPS trace gas data. In addition to model verification, the paper looks in detail at the forecast CO plume structure and evolution and seeks to provide a realistic explanation for the time-varying CO observations at Cape Grim.

In the next section a brief description of the AAQFS is given, with comments on the limitations of long-range transport in the AAQFS. We then present AAQFS results for the three distinct periods and compare these results with the CO observations at Cape Grim. The implications of the results presented are discussed, and concluding remarks are made.

AAQFS

The AAQFS consists of five major components: a numerical weather prediction (NWP) model (the Australian Bureau of Meteorology's Limited Area Prediction System, LAPS, Puri et al. (1998)), an emissions inventory module (Ng et al. 2000), a chemical transport module (CTM) for air quality modelling, an evaluation module, and a data archiving and display module. (Both LAPS and the CTM are Eulerian models.) For both the Victorian and New South Wales (NSW) domains the meteorology is modelled with a 0.05° horizontal grid and 29 vertical levels (nested in a 0.375° limited area model, which in turn is nested in a global model). The chemistry is modelled with a 0.01° horizontal grid with 17 vertical levels (Grid 2, covering urban areas) which is nested in a 0.05° horizontal grid (Grid 1, covering the full domain including rural areas). In the current study the operational Victorian Grid 1 domain was extended by 0.5° southward to include Cape Grim. The topographical map presented in Fig. 1 illustrates the coverage of this extended Grid 1 (Grid 2 results are not presented in this paper since they offer no further insight into the long-range transport). Tests on a Grid 1 domain that extended an additional 1° further south indicated that for the outflow conditions considered in this paper the results were unaffected by lateral boundary effects. A more detailed description of the system can be found in Cope et al. (2003). In addition to statistical verification, the performance of the AAQFS has been examined through a number of case studies. During the development of the system a number of verification studies were performed to highlight potential weaknesses and identify where improvements could be made (Cope et al. 1999; Hess et al. 2000a; Hess et al. 2000b and Tory et al. 2000). A statistical analysis of the system performance during the 2000/2001 photochemical smog season and the 2001 winter particle season is given in Cope et al. (2003); two photochemical smog case studies in Sydney and Melbourne are presented in Hess et al. (2003) and Tory et al. (2003), respectively.

Long-range transport

In the case studies mentioned above little emphasis was placed on long-range transport, and no long-range transport verification was attempted. There have been a number of occasions when long-range ozone transport in excess of 200 km has been present in both the Victorian and NSW forecast domains, although concentrations were never particularly high. This long-range ozone transport is difficult to verify, and is thus of limited use for assessing the AAQFS performance. Typically the ozone transport occurred above the surface, since surface ozone tends to be destroyed by near-surface emissions of nitric oxide and sulphur dioxide, and by redox reactions with surface objects at night. Verification of elevated ozone in the current network of monitoring stations, is not possible since they are all surface stations. Verification is further complicated by the fact that routine ozone monitoring is usually limited to major population centres. Thus, unless the ozone is carried from one centre to another and it arrives at the remote centre at night when local ozone concentrations are low, the remote ozone will not be identifiable.

Despite these limitations anecdotal evidence of rising ozone concentrations appearing in Canberra at night does support the long-range ozone transport found in the AAQFS forecasts. Formal studies are yet to be carried out to investigate this phenomenon. Hess et al. (2003) comment on the occurrence of AAQFS forecast ozone transport from Sydney to the Canberra region on two of the seven days of their study. In each case concentrations were relatively high even at the surface – pockets exceeded 80 ppb. (In this paper ppb refers to parts per billion by volume.) On both days the forecast ozone narrowly missed Canberra during the night (within about 50 km), although on one of the days elevated ozone did extend over Canberra in the early hours of the morning, and was mixed down to the surface when the mixed-layer deepened during the day (fumigation).

Examples of pollutant species transported over large distances in the AAQFS include bushfire smoke and wind-blown dust (dust storms). These events do provide opportunities for long-range transport verification in the AAQFS, but they are also subject to limitations. For the case of dust storms, a wind-blown dust module has only recently been introduced to the AAQFS (Lu and Shao 2001). It is dependent on vegetation type and cover, soil type, soil moisture and meteorological variables. Currently, the initialisation of the non-meteorological components in this module introduces a level of uncertainty that would not allow quantitative assessment of long-range transport for dust storm cases, although preliminary tuning of a number of parameters has shown promising results.

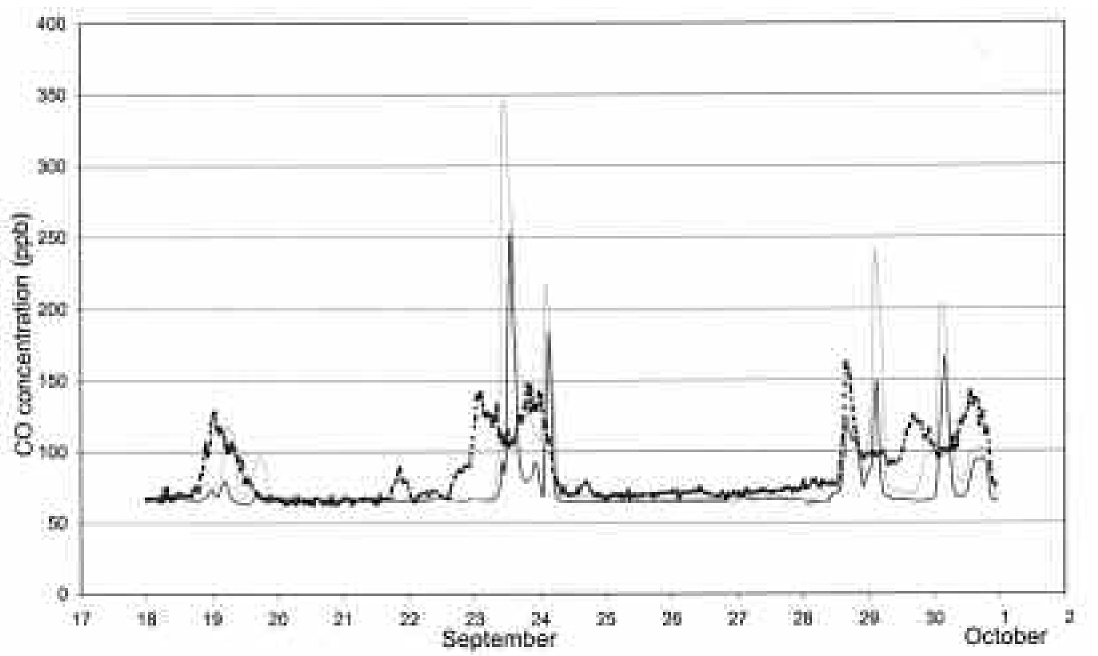
Limitations associated with using large bushfire events to assess long-range transport in the AAQFS include the necessity for a fire to develop a suitable distance upwind of monitoring stations. Although such events occur infrequently, the King Island bushfire, which transported thick smoke to Melbourne on the morning of January 11 2001, was an excellent example (Lee et al. 2001; Wain and Mills 2001). A fire that had been smouldering for a number of days on King Island flared up again after a cold front arrived from the southwest. The AAQFS successfully captured the position and structure of the smoke plume as it swept across Melbourne, and managed to predict a second peak in Melbourne, after the smoke had initially cleared at the surface, due to fumigation. However, the verification in this study was largely qualitative, since verification outside the Melbourne monitoring stations was provided by visual comparisons of modelled smoke plumes against satellite images. Modelling uncertainties in the quantity and height at which smoke (tracer particles) were released made quantitative assessment of concentrations difficult. A series of numerical experiments (Lee et al. 2001) demonstrated that the smoke transport to Melbourne (200 km away) was highly sensitive to the release height. This sensitivity arose largely because of the vertical variation in structure of the cold front that transported the smoke to Melbourne. Further quantitative uncertainties in that study were due to the inability to distinguish between smoke particles, sea-salt, vehicle emissions, dust and other particle sources in the measurements at the monitoring stations.

In this study uncertainty regarding pollutant source concentrations does exist, and this uncertainty limits the accuracy of the quantitative analysis. Y.L. Ng, of the Environmental Protection Authority, Victoria (personal communication) suggested the uncertainty on a particular day could be as high as 50 per cent. (The uncertainty associated with the annual CO emission estimate is about 20 per cent. Day-to-day variability increases the uncertainty for a particular day.) Despite the source uncertainty, we show in this paper that a quantitative assessment of long-range transport between Melbourne and Cape Grim (about 300 km) is a viable and useful exercise.

Verification

The study period began on 18 September 2001 and ran for 13 days. In this time elevated CO concentrations were recorded at Cape Grim during three distinct periods: 18-19 September, 23–24 September, and 28 September to 1 October. This is illustrated in the dashed line of Fig. 2. (This time-series comprises mean CO concentrations over a 40 second sampling

Fig. 2 **Time-series of observed CO concentrations (ppb) at Cape Grim (dashed line) and forecast maximum CO concentrations within a 0.1° (full dark line) and 0.5° (full pale line) radius of Cape Grim for the 14-day period, 18 September to 1 October, 2001. Vertical axis, CO concentration (ppb). Horizontal axis, time (days of the month).**



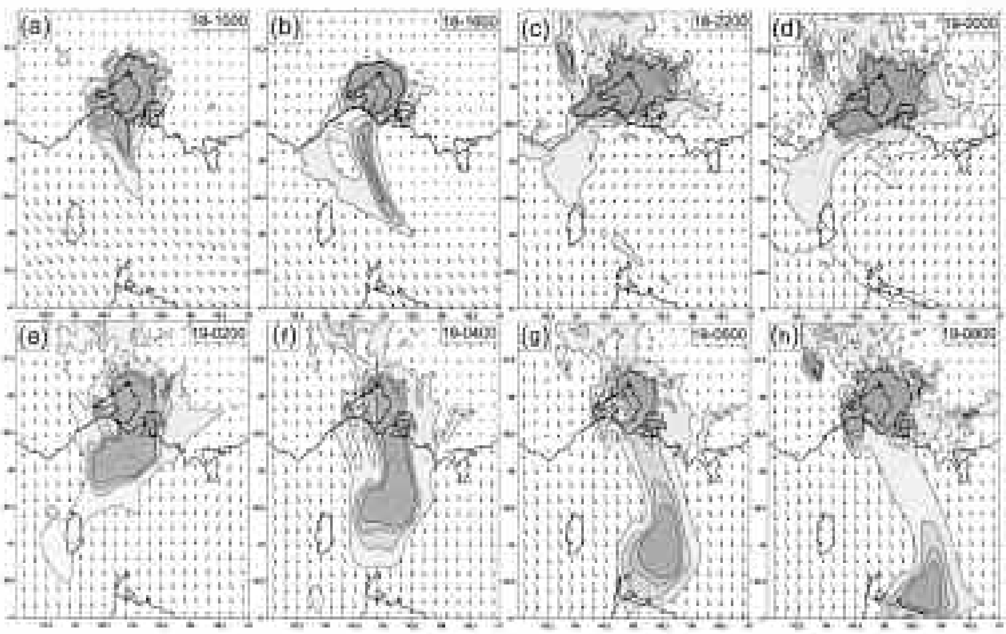
period, measured at 40 minute intervals.) The dark and pale full lines in Fig. 2 represent the maximum instantaneous modelled CO concentration within a 0.1 and 0.5° radial distance of Cape Grim respectively (i.e., the maximum instantaneous value at any grid point located within about 10 or 50 km of Cape Grim respectively). Since each of the three periods extends beyond the 36-hour AAQFS run time, the first 24 hours of successive runs have been used to provide a continuous record during these periods. (The AAQFS is run in a 'warm start' mode, in which the emissions from a previous forecast period are used to initialise the next forecast.)

An initial qualitative assessment of the system performance pertaining to Fig. 2, suggests it performed quite well with greater than background CO concentrations recorded during all three periods of observed elevated CO concentrations. The background CO concentration used in the system was 60 ppb – chosen to best match observed CO concentrations after the Period 1 plume had passed beyond Cape Grim. Thus, any significant increase above this level in the AAQFS record at Cape Grim can only be due to transport from the Melbourne airshed. (Other less significant CO sources and sinks in the AAQFS include CO production by photolysis of formaldehyde and decomposition of alkenes, CO destruction by reactions with hydroxyl radicals to form carbon dioxide, and the relatively insignificant Latrobe Valley source mentioned in the previous section.) A closer inspection of Fig. 2 shows that within a 0.1° radius of Cape Grim the magnitude of the first period maximum was significantly under-represented, and errors in the timing of CO peaks were present during the second and third periods, although the timing of the third period onset and cessation was very good (accurate to within an hour). Typical errors for trajectories, based on forecast winds, are of the order of 30 per cent or more of the travel distance (Stohl 1998), i.e., approximately 100 km or 1° for this study. Given this level of uncertainty of the forecast winds, an expectation of the plume to pass within a 0.1° radius (about 10 km) of Cape Grim to be considered a 'hit' is a particularly rigorous test of the system performance. Nevertheless, using the definition of a 'hit' as a period of above background CO concentration, Fig. 2 shows that the system passed this rigorous test during all three periods. The first period 'hit' was nearly a miss, given that the modelled plume just brushed past Cape Grim. Maximum values of 79, 87, 98, 111, 119 ppb passed within 0.1, 0.2, 0.3, 0.4 and 0.5° of Cape Grim respectively (only the 0.1 and 0.5° radii are represented in Fig. 2), compared to the first period maximum observed CO concentration of 127 ppb. The plume brushing past Cape Grim is illustrated in the sequence of images presented in Fig. 3.

Recognising that the use of a 0.1° detecting radius was a very stringent test of the system, time-series of maximum modelled CO concentrations within 0.2, 0.3, 0.4 and 0.5° detecting radii were also constructed to see if the system showed better quantitative performance when the rigorous detection procedure was relaxed. As expected, the additional time-series showed increases in peak CO concentrations with increasing detecting radius, due to more of the plume being sampled for the maximum value, and increases in CO peak widths, due to the plume entering the larger sample radius earlier and leaving later. Compared with the observed peaks, Fig. 2 shows the modelled peaks were sharper and of significantly greater amplitude, suggesting the modelled horizontal diffusion for CO may have been too small. Increasing the horizontal diffusion would lead to broader peaks with reduced magnitude in the modelled time series. This would lead in general to better agreement with the observations, with the exception of under-represented peak magnitude during the first period. At present the AAQFS contains no explicit horizontal diffusion in the CTM, only numerical diffusion. Experiments to examine the role that model diffusion plays in AAQFS will be the subject of a future paper.

In their assessment of the performance of TAPM, Cox et al. (1999) compared the period of time in which their model plume passed over Cape Grim with the observed time-period of elevated CO. A 'hit' was declared if there was an overlap between the two time-periods. They repeated this assessment for cases where the plume passed within 40 and 100 km boxes centred on Cape Grim. Of the 43 pollution episodes they identified during the 18-month period, their model plumes passed directly over the Cape Grim grid point during about 70 per cent of the events. The TAPM plumes passed over the 40 km square box on all but 2 events, and passed over the 100 km box during every event. These statistics suggest that if the AAQFS is performing as well as TAPM it would be expected to record a 'hit' during all three periods when a sample radius of 0.2° (about 20 km) was used. After subtracting the background CO concentrations Cox et al. (1999) recorded the ratio of observed to modelled CO concentrations, and made note of whether the edge or the centre of the modelled plume passed over Cape Grim. On those occasions when the edge of the modelled plume passed over Cape Grim the average ratio was about seven, and when the plume passed directly over Cape Grim the ratio was closer to three (i.e., the observed above background CO concentration was 7 and 3 times higher than modelled values). This result is consistent with the magnitudes recorded in Period 1 in the current study, when the edge of the modelled plume impacted on Cape

Fig. 3 CO (shaded, 80—140+ ppb, contour interval 20 ppb; additional 70 ppb contour in (d)) during Period 1 at (a) 1000, (b) 1600, (c) 2200 AEDT, 18 September, then at two-hour intervals until (h) 0800 AEDT, 19 September 2001. Forecast winds are indicated by vectors. The day of the month and time (AEDT) are included in the top right corner of each image.



Grim. However, in Periods 2 and 3, when the plume passed over Cape Grim, this ratio ranged from about 0.3 to 2. This shows that the AAQFS performance over the three periods studied was comparable to the TAPM performance averaged over 43 events.

In the remainder of this section the temporal and spatial distributions of the AAQFS forecast plumes are examined in detail, and a more quantitative assessment of the system performance is presented.

Period 1: 18 – 19 September 2001

The first-period CO-plume development and evolution is illustrated in Fig. 3, which shows the surface CO distribution between 1000 Australian Eastern Daylight Time (AEDT), 18 September and 0800 AEDT, 19 September 2001. Wind vectors are included to illustrate the direction of the winds responsible for shaping and transporting the modelled CO plume. An interesting feature of the sequence of images presented in Fig. 3, and also in Figs 4 and 5, is the appearance of CO 'pulsing' out of the Melbourne airshed due to high CO production during the morning and evening heavy commuting periods. This is best illustrated in Fig. 3(c) to (h), where the time interval between frames is two hours. The evening 'pulse' of 18 September drifted slowly to the southwest (Fig. 3(c),(d)), before a wind shift to northerly approached from the west, produced a kink in the plume and carried the CO across Bass Strait (Fig. 3(e),(f)). Between 0600 and 0800 AEDT 19 September 2001, the morning CO concentrations were building in the Melbourne airshed (Fig. 3(g),(h)), providing another 'pulse' to the plume, distinct from the previous evening pulse that brushed past Cape Grim.

The first instance of rising CO concentrations measured at Cape Grim began at about 2100 AEDT 18 September. These concentrations peaked at 0100 AEDT 19 September and returned to background levels by 1100 AEDT that day; there were fourteen hours of elevated concentrations in total. The panels presented in Fig. 3 illustrate the AAQFS plume over a similar period of time. The morning CO 'pulse' of 18 September had been advected southward and eastward, more than 100 km from the Melbourne airshed, by 1000 AEDT (Fig. 3(a)). Six hours later the plume was stretched and sheared by northeasterly winds in central Bass Strait that were maintained during the period and the development of onshore winds nearer the Victorian coast, where a sea-breeze circulation developed (Fig. 3(b)). This stretching and shearing is likely to have weakened the CO concentration through dilution.

Another six hours later, at 2200 AEDT, remnants of the modelled plume are just visible in a line extending from the northern tip of King Island towards the point where the southern boundary meets the 146° longitude (Fig. 3(c)). An extrapolation of this remnant plume, as it passed Cape Grim, is depicted in Fig. 3(d) by the addition of an extra contour line at 70 ppb. This gives an indication of the timing of an AAQFS plume arrival at Cape Grim, that is, the modelled plume of elevated CO concentrations arrived three hours later than the observed plume. If the plume had been more significant it is likely that it would have been broader as well as more highly concentrated, and hence its arrival at Cape Grim would then have been earlier and possibly more consistent with the observed plume arrival.

Another possible source of discrepancy between modelled and observed CO concentrations is wind error. Observation sites are chosen so that wind measurements best represent the larger scale local flow, by choosing sites with minimal influence from nearby small-scale features. Wind direction measurements tend to be more representative of the larger scale flow than wind speed measurements, due to the considerably greater influence of surface roughness on wind speed than wind direction. (The local surface roughness at the observation site can vary greatly from the average roughness of the general area that effects the larger-scale flow.) For this reason the discussion on wind errors will focus on wind direction rather than wind speed. In general, for the flow conditions experienced in this study, wind speed errors were larger at low wind speeds and smaller at the higher wind speeds when the plumes were being carried across Bass Strait; errors were often less than 10 per cent when the wind speed exceeded 6 – 7 m s^{-1}.

Comparisons of the observed wind directions at Cape Otway, Wilsons Promontory, King Island and Cape Grim (see Fig. 1 for locations), with the modelled winds showed very good agreement (not shown) during the early stages of Period 1 (1000 to 2200 AEDT 18 September). This suggests errors in modelled CO concentrations at Cape Grim during the early stages of Period 1 are more likely to be associated with plume size and concentration rather than errors in plume location. Unlike Periods 2 and 3, where the addition of explicit horizontal diffusion is likely to improve the forecast, in Period 1 it would only further dilute the plume. The evidence presented in the above three sentences suggests that there was an under-estimation of the emissions and/or an over-estimation of the Planetary Boundary Layer (PBL) height – both leading to an under-estimation of the maximum concentration. Over the last few years we have observed a small bias towards over-estimation of the PBL height in the marine boundary layer (observations taken at Sydney Airport, a coastal site), but it is not large enough to explain the significant discrepancies presented here. However, excessive vertical mixing in the AAQFS at the plume source, which is largely over land could be responsible for a significant reduction in CO concentration from dilution through a deep layer. Unfortunately the availability of PBL data was limited during that time. However, PBL depth observations at Melbourne Airport (about 20 km NNW (inland) of Melbourne) were available during the day on 18 September 2001, and show the PBL depth was over-predicted by 50 per cent.

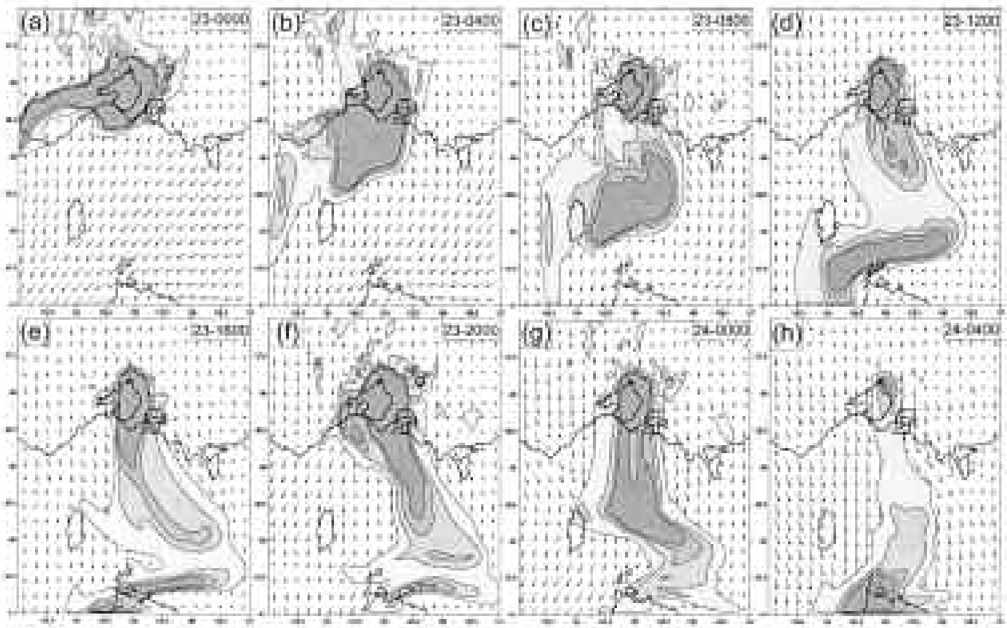
The remaining eight hours of plume development presented in Fig. 3 (e) to (h) has already been discussed (above) in terms of the CO plume 'pulse'. In the discussion it was noted that the plume 'brushed' past Cape Grim (Fig. 3(g) to (h)). Wind observations at King Island and Cape Grim show a NNE direction from 0400 to 0800 AEDT 19 September rather than the NNW direction of the modelled winds shown in (Fig. 3(f) to (g)). This suggests the near miss may have been due to an overly strong westerly component to the modelled wind.

In this study, observations are limited to wind measurement at only a few sites and CO concentration measurements at a single location (Cape Grim). However, it is interesting to note that a more substantial plume early in Period 1, as proposed above, is likely to have remained in the Cape Grim area for a number of hours after that shown in Fig. 3(d), and could have merged with the next plume 'pulse' pictured arriving in Fig. 3(f). Add to this a greater horizontal spread of the arriving plume to account for the suggested lack of horizontal diffusion in the system, mentioned earlier, and we have a scenario that could explain the more extensive period of elevated CO concentrations that occurred in reality (Fig. 2).

Period 2: 23 – 24 September 2001

Both the observed and modelled CO time series presented in Fig. 2 show double peaks during the second period, with the modelled peaks lagging the observed peaks by a number of hours. An examination of the observed winds at Cape Grim shows that these differences are not solely due to timing errors, as the first observed peak occurred during sustained easterly and ESE winds. This means the CO sources responsible for the first observed peak are likely to have included Burnie, an industrial town located about 100 km ESE of Cape Grim (just off the edge of Fig. 1) and local agriculture. CO sources outside the Melbourne airshed were not included in the AAQFS forecasts. The modelled double peak was caused by the arrival of successive Melbourne evening CO pulses at Cape Grim. Shifting winds were largely responsible for the

Fig. 4 Same as Fig. 3 except for Period 2. Four-hour time intervals separate each image ranging from (a) 0000 AEDT, 23 September to (h) 0400 AEDT, 24 September 2001.



clearing of CO between the pulses. The sequence of images presented in Fig. 4 illustrates the shifting winds over the Victorian coastal area, from NE to NW (Figs 4(a) to (d)) and then back to the north (Fig. 4(h)). During most of this time the winds over southern Bass Strait had a stronger easterly component. This combination of wind directions led to the plume folding evident in Fig. 4.

The first modelled CO peak that arrived at Cape Grim near midday on 23 September (Fig. 4(d)), resulted from the evening Melbourne CO pulse generated on 22 September. The CO plume was originally advected WSW (Fig. 4(a)) before NNW winds transported the plume on a relatively large front towards Cape Grim (Fig. 4(b),(c)). Convergence of the NNW winds with the NE winds in southern Bass Strait led to the twisting and elongation of the plume as it reached Cape Grim (Fig. 4(d)). Four hours later this elongated part of the plume had thinned considerably and was just clearing Cape Grim (Fig. 4(e)). A clear region followed before the next weak pulse (originating from the morning peak period, and corresponding to the small bump in the record between the two second-period peaks in Fig. 2) approached Cape Grim (Fig. 4(f)). Most of the morning pulse appears to have passed to the east of Cape Grim with only a rather diffuse edge reaching the site (Fig. 4(d) to (f)). The second modelled peak resulted from the 23 September evening pulse being advected almost directly southward to Cape Grim by strengthening northerly winds (Fig. 4(g) to (h)).

As mentioned above, the first observed peak during Period 2 has been attributed to westward CO transport from Burnie. Wind observations at Cape Otway, Wilsons Promontory, King Island and Cape Grim suggest that CO transport from Melbourne by northerly flow may have arrived at Cape Grim between 1200 and 1600 AEDT, which corresponds to the time that the second peak was building, in which case the first modelled peak arrived a few hours early and the magnitude was too strong. The observed winds at King Island and Cape Grim had a stronger easterly component than the AAQFS forecast winds from 2000 AEDT onwards (i.e., the model westerlies were too strong). This wind error may have been responsible for the gap between the two modelled CO peaks. Again, increased horizontal diffusion may spread the modelled plume further and lead to broader and less sharp peaks in better agreement with the observed CO time-series.

Period 3: 28 – 30 September 2001

Period 3 is characterised by multiple CO peaks in both the observed and modelled CO time-series (Fig. 2).

The sequence of images presented in Fig. 5 shows predominantly northerly flow throughout the period with a shifting zonal component from east to west to east and then back to west again. This changing wind pattern was responsible for the sweeping back and forth of the modelled plume across Bass Strait and ultimately across Cape Grim. The first modelled peak was caused by downward mixing of greater CO concentrations aloft to the surface (fumigation). Stronger northerly winds above the surface produced a plume that extended about 50 to 100 km further south than the surface plume. Figure 5(a) shows the surface plume at midday approaching Cape Grim about four hours before fumigation occurred. Four hours later an isolated region of CO appeared over Cape Grim (Fig. 5(b)) and remained separate from the main plume for at least another four hours (Fig. 5(c)). The fumigation is illustrated in Fig. 6, which shows a diagonal vertical section passing through King Island and Cape Grim (See Fig. 1 for the diagonal section location). An hour before the fumigation occurred a region of elevated CO (between 300 and 600 m above the surface) had just passed over the Cape Grim area (Fig. 6(a)). By 1600 AEDT, Fig. 6(b) shows that the CO had mixed down to the surface. The forecast winds compared well with the observations at all stations during this time. This agreement combined with the very good timing and duration of the first model peak, suggests that the fumigation explanation of the peak is quite plausible. Alternatively a more vertically uniform plume could also produce a peak consistent with that observed.

The shift from NE to NW flow that was responsible for the second modelled peak at Cape Grim is first evident between Cape Otway and King Island in Fig. 5(d). It had transported the plume to Cape Grim by 0400 AEDT 29 September (Fig. 5(e)). A period of about 20 hours then followed when the plume continued to sweep eastward beyond Cape Grim before being carried back by another shift in wind direction (Fig. 5(f) to (k)). The observed winds showed the first modelled wind shift arrived about 8 to 12 hours early and the second about 8 hours late. It is likely that a plume transported by the observed winds would have arrived from the west 8 to 12 hours later than the modelled plume, and then would have been advected back to the west before it had a chance to pass beyond Cape Grim. In other words, the second and third modelled peaks would have been brought backward and forward in time respectively to merge into one peak, consistent with the second observed peak present in Period 3 of Fig. 2.

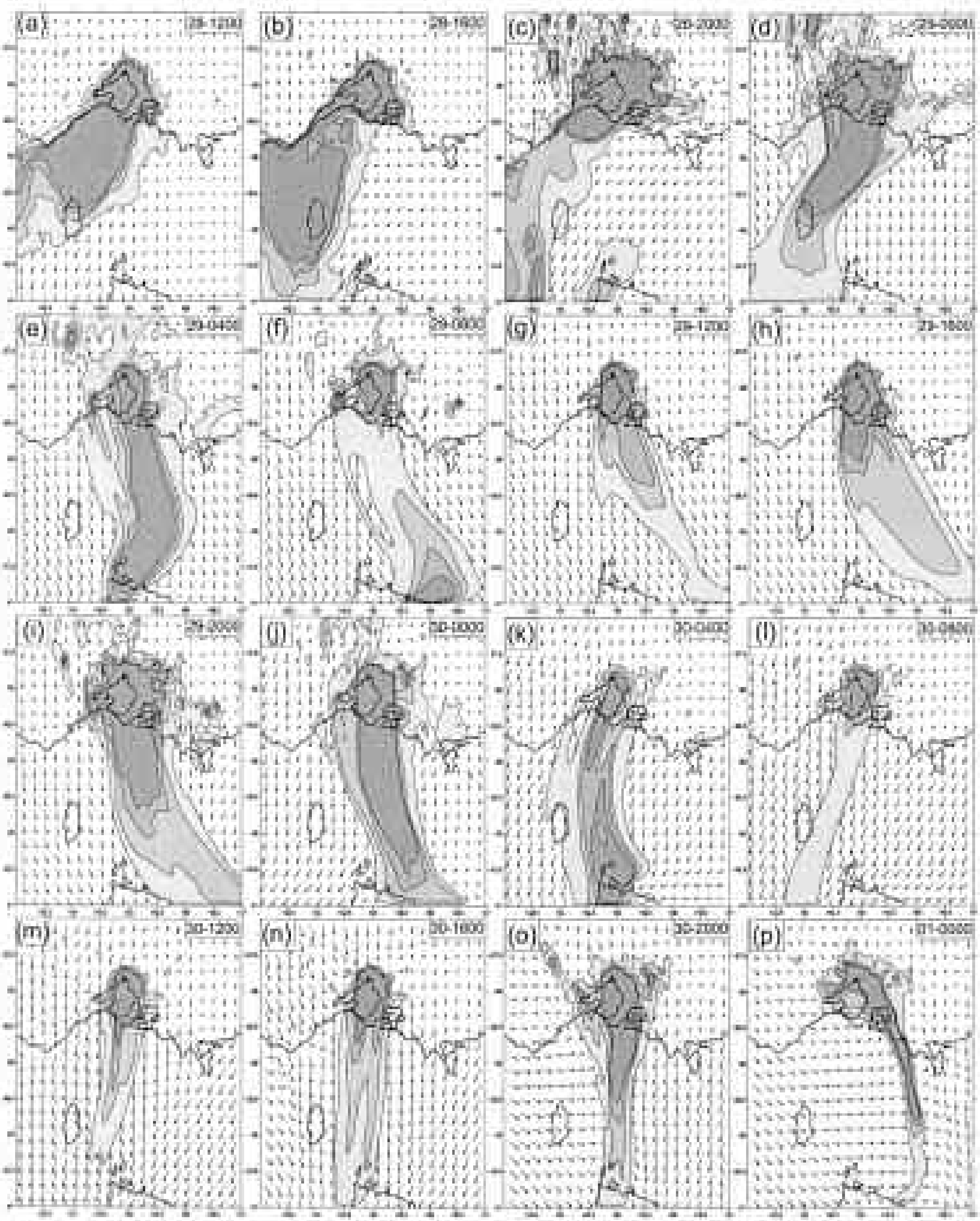
The fourth modelled peak of Period 3 resulted from the final east to west shift of the zonal component of the wind caused by the passage of a cold front (Fig. 5(l) to (p)). The observed winds during this time showed that the timing of the front was well forecast (accurate to within a couple of hours). The timing of the final clearing of the modelled CO is closely coincident with the observed CO clearing, which further supports the accuracy of the frontal passage. The modelled plume was particularly weak during this period possibly due to over-dilution in the vertical. The system is slightly biased towards over-predicting the mixed layer depth over the sea (Hess et al. 2003). This could partly explain the weaker and later arriving modelled plume. It is interesting to note how the plume was influenced by the passage of the front. Leading the front (defined by the sharp change to a westerly wind direction) is a wedge of northerly flow in the shape of an upright V (see Fig. 5(n), where the 'V' runs roughly from Cape Otway to Cape Grim and up to Wilsons Promontory). Ahead of these northerly winds is the preceding background NE flow. The final modelled CO peak arrived with this 'V' of northerly winds (Fig. 5(n)). Four hours later the front had reached Cape Grim and the convergence within the frontal leading edge had begun to contract the east-west scale of the plume, and increase the east-west CO concentration gradient (Fig. 5(o)). Another four hours later the CO plume had been compressed into a very narrow line across Bass Strait and advected eastward beyond Cape Grim (Fig. 5(p)). Figure 5(p) shows the front leading the CO plume. This suggests that the front propagated through Bass Strait like a wave rather than like a gravity current and hence the plume was not transported with the leading edge of the front. Vertical east-west profiles through the front (not shown) revealed the front propagated through the plume at all levels, not just the surface.

Another factor that could contribute to the separation of the front from the plume is related to the way in which the CTM uses the meteorological data from LAPS. At the completion of every hour of integration the meteorological fields are taken from LAPS and used to drive the CTM for the next hour. In each one-hour period these fields are constant. Thus, the CTM sees sudden changes in wind speed and direction every hour, and it is possible that the front 'jumped' through the plume to some extent. If this was the sole cause of the separation between the plume and frontal leading edge, one might expect a maximum of a one-hour separation. Estimates of frontal translation speed from Fig. 5(o) and 5(p) (four-hour interval) would suggest the separation is larger than one hour.

Discussion and conclusions

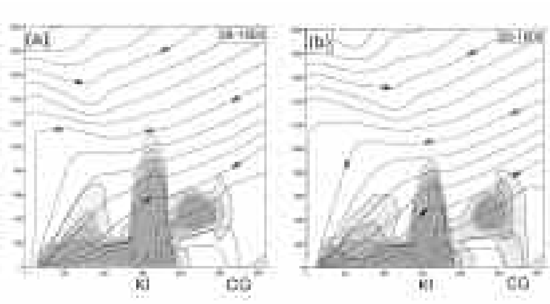
As a forecasting tool, the AAQFS has demonstrated considerable success in identifying 'pollution' events, at a qualitative level, during this 13-day period. If a 'hit' is defined as a period in which the modelled CO

Fig. 5 Same as Fig. 3 except for Period 3. Four-hour time intervals separate each image ranging from (a) 1200 AEDT, 28 September to (h) 0000 AEDT, 01 October 2001.



concentration is greater than the background level then the system achieved a 100 per cent success rate. A more quantitative assessment of the modelled CO time-series at Cape Grim in the previous section identified errors in the timing of CO peaks and the duration and magnitude of the modelled CO concentrations. Comparisons between the modelled and observed winds identified trajectory errors as the most likely cause of the timing errors in the Cape Grim CO time-series, and it was proposed that insufficient horizontal diffusion may be responsible for the too brief and intense CO peaks in the Cape Grim time-series. Potential CO source errors could effect the 'peakiness' of the CO time-series, but would have

Fig. 6 Vertical cross-sections passing through King Island (KI) and Cape Grim (CG), illustrating fumigation at Cape Grim. See Fig. 1 for the cross-section location. Potential temperature is contoured (interval 1K) and CO is shaded (shade increments identical to Figs 3 to 5) at (a) 1500 and (b) 1600 AEDT, 28 September 2001. Vertical and horizontal scale, metres and kilometres respectively.



little effect on the duration of the elevated CO record at Cape Grim. Experiments with diffusion are likely to lead to improvements in the shape of the peaks in the single point time-series, which should lead to a more realistic plume distribution. These experiments will be performed in a future study. However, the more significant errors present in the time-series are associated with trajectory errors.

Trajectory errors are not only associated with wind direction errors but also errors in the timing of wind changes. It might be expected that larger scale (i.e., synoptic) winds may be mostly responsible for long-range transport of pollutants, since sustained periods of nearly constant winds over a significant area are required to transport pollutants in elevated concentrations over long distances. The direction of large-scale winds and the timing of large-scale wind changes, are mostly dependent on the large-scale mass distribution within the model atmosphere. This mass distribution is in turn dependent on the fundamental make-up of the NWP model and the model initialisation. Uncertainties in the model initial conditions will always be present. This uncertainty, combined with the chaotic nature of the atmosphere, means trajectory errors that grow with time are unavoidable. The components of the model that have the greatest uncertainty, outside of the initialisation, such as the land-surface or planetary boundary layer schemes, have only a secondary influence on the large-scale mass distribution and hence synoptic-scale winds. Thus, development in these areas can only improve the forecast winds up to a point.

Single-point performance assessment of the AAQFS through time-series comparisons alone provides a rigorous test of the system, and helps to identify the limits of long-range predictability. Figure 2 shows that timing errors of the order of 12 hours can be expected from this system for long-range transport over distances of about 300 km. However, greater insight can be obtained from the AAQFS by examining the spatial and temporal distribution of the plume. In hindcast mode (i.e., running the forecast after the event), such as in this study, a plausible picture of the true CO plume evolution and transport can be constructed using the AAQFS as guidance when interpreting the available observations. As a forecast tool a good indication of the plume structure and transport can be obtained using the AAQFS as guidance.

Features such as the CO pulses, plume folding, fumigation, sweeping of the plume back and forth across Bass Strait, and the modification of the plume by the cold front, are all examples of interesting plume behaviour identified by the AAQFS. Although a greater distribution of observing sites would be necessary to verify these behaviours in detail, they provide insight into aspects of long-range transport of potential importance between Melbourne and Cape Grim, and in other regions of the globe where long-range transport is known to be important.

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References

Cope, M., Hess, D., Lee, S., Azzi, M., Carras, J., Wong, N. and Young, M. 1999. Development of the Australian Air Quality Forecasting System: current status. *Proceedings of the International Conference on Urban Climatology,* Sydney, 8–12 November 1999.

Cope, M.E., Hess, G.D., Lee, S., Tory, K.J., Azzi, M., Carras, J., Lilley, W., Manins, P.C., Nelson, P., Ng, L., Puri, K., Wong, N., Walsh, S. and Young, M. 2003. The Australian Air Quality Forecasting System. Part I. Project description and early outcomes. *Jnl appl. Met.* (in press).

Cox, M., Hurley, P., Fraser, P. and Physick, B. 1999. Investigation of Melbourne region pollution events using Cape Grim data, a regional transport model (TAPM) and the EPA Victoria carbon monoxide inventory. *Clean Air, 33*, 35-40.

Fennetaux, I., Colin, P., Etienne, A., Boudries, H., Dutot, A.L., Perros, P.E. and Toupance, G. 1999. Influence of continental sources on oceanic air composition at the eastern edge of the North Atlantic Ocean, TOR 1992–1995. *J. Atmos. Chem., 32*, 233-80.

Hess, G.D., Cope, M.E., Lee, S., Manins, P.C., Mills, G.A., Puri, K., Tory, K. 2000a. The development of the Australian Air Quality Forecasting System: current status. *Proceedings of the Millennium NATO/CCMS International Technical Meeting on Air Pollution Modelling and its Application*, 15–19 May 2000, Boulder CO, American Meteorological Society, 276-83.

Hess, G.D., Cope, M.E., Lee, S., Manins, P.C., Mills, G.A., Puri, K. and Tory, K. 2000b. The Australian Air Quality Forecasting System. *AMOS Bulletin, 13*, 67-73.

Hess, G.D., Tory, K.J., Cope, M.E., Lee, S., Puri, K., Manins, P.C. and Young, M. 2003. The Australian Air Quality Forecasting System. Part II. Case study: Sydney 7-day event. *Jnl appl. Met.* (in press).

Jaffe, D., Anderson, T., Covert, D., Kotchenruther, R., Trost, B., Danielson, J., Simpson, W., Berntsen, T., Karlsdottir, S., Blake, D., Harris, J., Carmichael, G. and Uno, I. 1999. Transport of Asian air pollution to North America. *Geophys. Res. Lett., 26*, 711-14.

Kallos, G., Kotroni, V., Lagouvardos, K. and Papadopoulos, A. 1998. On the long-range transport of air pollutants from Europe to Africa. *Geophys. Res. Lett., 25*, 619-22.

Kato, S., Pochanart, P. and Kaji, Y. 2001. Measurement of ozone and nonmethane hydrocarbons at Chichi-juna Island, a remote island in the western Pacific: Long-range transport of polluted air from the Pacific rim region. *Atmos. Environ., 34,* 6021-9.

Lee, S., Cope, M., Tory, K., Hess, D. and Ng, L. 2001. The Australian Air Quality Forecasting System: Modelling of a severe smoke event in Melbourne, Australia. *Proceedings of the 25th NATO/CCMS International Technical Meeting on Air Pollution Modelling and its Application*, 15-19 October 2001, Louvain-la-Neuve, Belgium.

Lu, H. and Shao, Y. 2001. Toward quantitative prediction of dust storms: an integrated wind erosion modelling system and its applications. *Environmental Modelling and Software, 16,* 233-49.

Manins, P.C. (Chair of Committee) 2001. *Air Quality Forecasting for Australia's Major Cities – Final Report*. Project Management Committee: CSIRO Atmospheric Research, Aspendale, Australia: http://www.dar.csiro.au/info/aaqfs/

Ng, Y.L., Walsh, S. and Wong, N. 2000. Emissions module for the Australian Air Quality Forecasting System. *Proceedings 15th International Clean Air and Environment Conference*, Sydney, 275–80.

Puri, K., Dietachmayer, G., Mills, G.A., Davidson, N.E., Bowen, R.A. and Logan, L.W. 1998. The new BMRC Limited Area Prediction System, LAPS. *Aust. Met. Mag., 47*, 203-33.

Stohl, A. 1998. Computation, accuracy and application of trajectories - a review and bibliography. *Atmos. Environ., 32,* 947-66.

Tory, K.J., Hess, G.D., Mills, G.A. and Puri, K. 2000. Verification of the meteorological component of the Australian Air Quality Forecasting System. *Proceedings 15th International Clean Air and Environment Conference*, Sydney, 221-6.

Tory, K.J., Cope, M.E., Hess, G.D., Lee, S., Puri, K., Manins, P.C. and Wong, N. 2003. The Australian Air Quality Forecasting System. Part III. Case study: Melbourne 3-day event. *Jnl app. Met.* (in press).

Wain, A. and Mills, G. 2001. Smoke dispersion forecasting for prescribed burning: a status report. *Proc. Bushfire 2001 - Australasian Bushfire Conference*, 3-6 July 2001, Christchurch, New Zealand.

Wanger, A., Peleg, M., Sharf, G., Mahrer, Y., Dayan, U., Kallos, G., Kotroni, V., Lagouvardos, K., Varinou, M., Papadopoulous, A. and Luria, M. 2000. Some observational and modeling evidence of long-range transport of air pollutants from Europe toward the Israeli coast. *J. Geophys. Res., 105*, 7177-86.