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Modelling horizontal and vertical concentration profiles of ozone and oxides of nitrogen within high-latitude urban areas

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Abstract

Urban ozone concentrations are determined by the balance between ozone destruction, chemical production and supply through advection and turbulent down-mixing from higher levels. At high latitudes, low levels of solar insolation and high horizontal advection speeds reduce the photochemical production and the spatial ozone concentration patterns are largely determined by the reaction of ozone with nitric oxide and dry deposition to the surface. A Lagrangian column model has been developed to simulate the mean (monthly and annual) three-dimensional structure in ozone and nitrogen oxides (NO_x) concentrations in the boundary-layer within and immediately around an urban areas. The short-time-scale photochemical processes of ozone and NO_x , as well as emissions and deposition to the ground, are simulated. The model has a horizontal resolution of 1×1 km and high resolution in the vertical. It has been applied over a 100×100 km domain containing the city of Edinburgh (at latitude 56°N) to simulate the city-scale processes of pollutants. Results are presented, using averaged wind-flow frequencies and appropriate stability conditions, to show the extent of the depletion of ozone by city emissions. The long-term average spatial patterns in the surface ozone and NO_x concentrations over the model domain are reproduced quantitatively. The model shows the average surface ozone concentrations in the urban area to be lower than the surrounding rural areas by typically 50% and that the areas experiencing a 20% ozone depletion are generally restricted to within the urban area. The depletion of the ozone concentration to less than 50% of the rural surface values extends only 20 m vertically above the urban area. A series of monitoring sites for ozone, nitric oxide and nitrogen dioxide on a north–south transect through the city – from an urban, through a semi-rural, to a remote rural location – allows the comparison of modelled with observed data for the mean diurnal cycle of ozone concentrations. In the city centre, the cycle is well reproduced, but the ozone concentration is consistently underestimated. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Tropospheric ozone; Lagrangian column model; Urban; Nitrogen oxides; Vertical exchange

1. Introduction

Tropospheric ozone is a photochemical oxidant formed largely by photochemical reactions. In the troposphere ozone acts as a greenhouse gas (Fishman et al., 1979; Chalita et al., 1996) and is toxic to plants, reducing

crop yields (Hewitt et al., 1990), and to humans as a respiratory irritant (WHO, 1987), as well as damaging both natural and man-made materials, such as stone, brickwork and rubber (PORG, 1993). Quantifying the dose and exposure of the human population, vegetation and materials to ozone is required to assess the scale of ozone impacts and to develop control strategies.

A network of 17 rural ozone-monitoring stations across the UK provides broad-scale regional spatial patterns in tropospheric ozone concentrations in rural areas (PORG, 1993). Peak ozone concentrations increase from north to south across the UK as the south has higher

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concentrations of the primary pollutants necessary for ozone production (from greater emissions and proximity to continental sources) as well as greater frequency of meteorological conditions suitable for ozone production. Mean ozone concentrations also increase with altitude (PORC, 1997) and are higher in a 5–10 km coastal strip (Entwistle et al., 1997). The mean annual-average background concentration of ozone for the UK is approximately $50 \mu\text{g m}^{-3}$, though there is a wide variation for individual episodes about this value, ranging up to about $400 \mu\text{g m}^{-3}$ (PORC, 1993). There are clear diurnal and annual cycles in ozone concentrations in the UK, with a mid-afternoon peak and nocturnal minimum and a spring maximum and autumn minimum. The diurnal cycle illustrates the fundamental importance of vertical mixing (Garland and Derwent, 1979).

Ozone concentrations in urban areas are of particular interest and importance as the population is largely urban-based and ozone concentrations show greater spatial and temporal variations in urban areas. Ozone concentrations are smaller in urban areas than they are in surrounding rural areas due to the reaction of ozone with nitric oxide, emitted from the combustion sources, forming nitrogen dioxide. Where air is allowed to stagnate over an urban area, the effects of strong insolation and accumulating ozone precursors can produce very high ozone concentrations. An example of this is the photochemical smog that affects the Los Angeles area of California, where a combination of meteorology, local topography and very high pollutant emission levels produce dangerously high ozone concentrations on many days of the year (Lents and Kelly, 1993). However, in most parts of the UK and other high-latitude areas where insolation levels are lower and wind speeds are larger (maintaining a steady advection of air through the urban air-shed), there is a different spatial distribution of concentrations with annual mean ozone concentrations generally smaller in urban areas.

There are many urban sites at which ozone is monitored in the UK (PORC, 1997). Attempts to map ozone concentrations using both rural and urban monitoring are complicated by the interaction of local chemistry with the larger-scale meteorological factors determining ozone concentrations. Recent studies on higher-resolution ozone mapping have used an urbanisation index (PORC, 1997), but these have not been able to account for the movement of ozone and ozone-depleted air into and out of the urban areas.

Various studies have examined the characteristics of ozone and nitrogen oxide concentrations around urban areas (Ball and Bernard, 1978). A large number of studies have focussed on the two-dimensional structure of the 'urban plume' of photochemical-ozone downwind of a city, with different cities around the world being studied (Cleveland et al., 1976; White et al., 1976; Varey et al., 1988; Lin et al., 1996; Silibello et al., 1998). There have

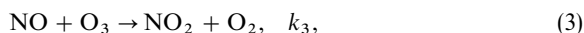
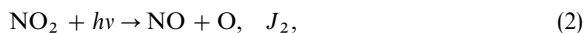
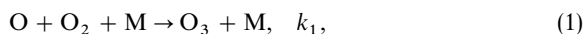
been few studies of the three-dimensional mean structure of ozone concentrations in urban areas, especially at high latitudes, where ozone production is of secondary importance in describing the spatial distributions around cities. These studies are largely interpretations of observational data (Angle and Sandhu, 1989; Leahey and Hansen, 1990).

A boundary-layer Lagrangian column model has been developed to simulate the mean three-dimensional structure in ozone and nitrogen oxide concentrations in the boundary-layer within and immediately around high-latitude urban areas at a spatial scale of $1 \times 1 \text{ km}$. (The model is not appropriate for use with "real-time" trajectories.) The model simulates the effects of ozone depletion at the surface as a consequence of the reaction with emitted nitric oxide and dry deposition to the surface. This has been used to follow a range of one-dimensional trajectories over a distance of 10^5 m and a travel time in the order of 10^4 s , through a simulated city under a variety of meteorological and pollutant emission regimes representing seasonal and diurnal extremes. An assessment of the extent of ozone destruction occurring, the rate of recovery of surface ozone concentrations downwind of the city and the influence of meteorological parameters on the ozone concentration has been provided using the model.

The model has been applied over a $100 \times 100 \text{ km}$ domain containing a simulation of the emission field over the city of Edinburgh. Edinburgh was used as a generic, high-latitude city for modelling purposes. A land-use array has been created as input to the model with spatially and temporally variable emission and deposition values.

2. Ozone destruction by nitric oxide

High-latitude cities are, in general, well ventilated, so that the timescale for air traversing the city is small compared to that for ozone generation. Under these conditions, the three reactions that are fundamental in the determination of ozone (O_3) concentrations in urban areas (Wayne, 1991) are



where k_1 and k_3 are reaction rate constants and J_2 is the photolysis rate for nitrogen dioxide (NO_2).

In the UK, the main source of nitrogen oxides is the combustion of fossil fuels, i.e. the burning of coal, oil and gas in power stations and the combustion of petrol and diesel by road traffic (Salway et al., 1997). The largest source in large urban areas is the exhaust from road

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