Environmental Pollution 188 (2014) 132–143

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Modelling segregation effects of heterogeneous emissions on ozone levels in idealised urban street canyons: Using photochemical box models

Jian Zhong, Xiao-Ming Cai*, William James Bloss

School of Geography, Earth & Environmental Sciences, University of Birmingham, Edgbaston, Birmingham B15 2TT, UK

ARTICLE INFO

Article history:
Received 9 December 2013
Received in revised form
1 February 2014
Accepted 3 February 2014

Keywords:
Segregation effect
Urban street canyon
Emission heterogeneity
Photochemical box model
Urban ozone concentrations

ABSTRACT

Air quality models include representations of pollutant emissions, which necessarily entail spatial averaging to reflect the model grid size; such averaging may result in significant uncertainties and/or systematic biases in the model output. This study investigates such uncertainties, considering ozone concentrations in idealised street canyons within the urban canopy. A photochemical model with grid-averaged emissions of street canyons is compared with a multiple-box model considering each canyon independently. The results reveal that the averaged, 'one-box' model may significantly underestimate true (independent canyon mean) ozone concentrations for typical urban areas, and that the performance of the averaged model is improved for more 'green' and/or less trafficked areas. Our findings also suggest that the trends of 2005–2020 in emissions, in isolation, reduce the error inherent in the averaged-emissions treatment. These new findings may be used to evaluate uncertainties in modelled urban ozone concentrations when grid-averaged emissions are adopted.

1. Introduction

Atmospheric chemical and physical processes are tightly coupled in air quality simulations (Karamchandani et al., 2012). A general operating hypothesis of most urban air quality grid-based models is that primary air pollutants emitted from vehicles, industry or other sources are instantaneously well-mixed or distributed within the entire model grid-cell which contains the emissions (Auger and Legras, 2007). The grid-averaged emission rates of primary air pollutants are normally used as an input representing the mean gridded emissions (Denby et al., 2011) in atmospheric chemical models and the concentration in the canopy layer is modelled as one box representing the canopy layer for the entire grid cell. However, in reality these surface emissions vary, and exhibit a high temporal and spatial heterogeneous distribution at the sub-grid scale, referred to as surface sub-grid emission heterogeneity (Galmarini et al., 2008). This leads to segregation effects due to incomplete mixing. In the grid-averaging procedure, all sub-grid scale processes and features (Ching et al., 2006) are lost and secondary pollutants (e.g. O_3) may therefore be systematically under- or over-estimated.

Several model approaches have been suggested to account for the impacts of sub-grid emission heterogeneity. Nested-grid or high-resolution modelling is a simple approach to resolve sub-grid scale variability. Examples of such approach can be seen from the Community Multiscale Air Quality (CMAQ) model (Sokhi et al., 2006; Shrestha et al., 2009), the Weather Research and Forecasting/Chemistry (WRF/Chem) model (Grell et al., 2005), and the Comprehensive Air Quality Model with extensions (CAMx) (Shen et al., 2011). A shortage of this approach is that it is only effective locally to a fixed area where the finer resolution grid is located. In order to overcome the limitation, adaptive grid modelling (Srivastava et al., 2000; Constantinescu et al., 2008; Garcia-Menendez et al., 2010) was developed to allow dynamic change of the grid system during a simulation. Garcia-Menendez and Odman (2011) discussed the details and reviewed the advances of the adaptive grid modelling. Another approach to incorporate sub-grid emission heterogeneity is hybrid modelling, which combines a regional grid-based model with a local Gaussian dispersion model (e.g. ADMS (Arciszewska and McClatchey, 2001) and AERMOD (Zou et al., 2010)). This approach has been extensively implemented, such as the CMAQ-ADMS model (Chemel et al., 2011; Beevers et al., 2012; Stocker et al., 2012), the CMAQ-AERMOD model (Stein et al.,

* Corresponding author.
E-mail address: x.cai@bham.ac.uk (X.-M. Cai).

http://dx.doi.org/10.1016/j.envpol.2014.02.001
0269-7491/

Nomenclature

$C_{i,m}$	Concentration of the i^{th} species in Box m ($m = 0,1,2$) (ppb);
$C_{bi,m}$	Background concentration of the i^{th} species for Box m ($m = 0,1,2$) (ppb);
$C_{i,1+2}$	Averaged concentration of Boxes 1 and 2 of the i^{th} species (ppb);
$E_{i,m}$	Emission rate of the i^{th} species in Box m ($m = 0,1,2$) (ppb s^{-1});
H_m	Height of the street canyon of Box m ($m = 0,1,2$) (metre);
$I_{S(A+B)}$	Intensity of segregation between species A and B;
$k_{(A+B)}$	Second-order rate constant for species A and B in a well-mixed box;
$\langle k_{eff\,(A+B)} \rangle$	Effective second-order rate constant for species A and B in the 'two-box' model;
RSL	Region Split Line;
t	Time (s);
$w_{t,m}$	Exchange velocity between street canyon and background for Box m ($m = 0,1,2$) (m s^{-1});
$\Delta S_{i,m}$	Net chemical production rate of the i^{th} species in Box m ($m = 0,1,2$) (ppb s^{-1});
ε	Heterogeneity of emissions;
ϕ_i	Percentage of overestimation for the i^{th} species by the 'one-box' model (%);

2007; Isakov et al., 2009; Johnson et al., 2010) and the WRF-AERMOD model (Kesarkar et al., 2007). A more promising approach is the plume-in-grid (PinG) modelling (Karamchandani et al., 2002), which imbeds a non-steady-state plume model inside the grid. Vijayaraghavan et al. (2006) implemented the plume-in-grid (PinG) modelling approach in the CMAQ-APT model to reduce sub-grid scale variability in a simulation of central California. They found that the sub-grid treatment can lead to up to 10 ppb less O_3 under the condition of O_3 formation and up to 6 ppb more O_3 under other conditions, compared with a base simulation without the PinG treatment. The approach offers a more realistic representation of the elevated point emission sources and their atmospheric fate. Galmarini et al. (2008) developed a Reynolds-average model to parameterize sub-grid emission heterogeneity in the meso- and global scale. Their study built upon the assumption that concentrations can be divided into a mean part, depending upon the average emissions, and a fluctuation component which depends on the variability of emissions, respectively. Alternatively, Cassiani et al. (2010) developed a stochastic fields method to address surface sub-grid emission heterogeneity in a mesoscale dispersion model. The advantage of this method is that the sub-grid scale emission variability is well-represented by the probability density functions. Some of the above approaches to address sub-grid scale errors are also reviewed and discussed in details by Touma et al. (2006) and Karamchandani et al. (2011). Currently, strategies to address sub-grid emission heterogeneity are mostly focussed upon large scale grid-based models. However, for the small scale, there is little research focussing on the effects of sub-grid emission heterogeneity.

Here, we extend consideration of emissions heterogeneity to the small scale, i.e. the canyon scale. The canopy layer is a major source for emissions into the overlying atmosphere/boundary layer and is normally within the lowest grid-cell of a grid-based model. From the canopy layer perspective, urban street canyons are typical sub-grid scale features separated by rows of buildings. These emissions into the canyon layer may be pre-processed within urban street canyons before they enter to the entire grid-cell in the lowest part of the grid-based model (Fisher et al., 2006). Urban street canyons, where human exposure takes place, are the area of interest in this paper. The additional information between the grid-averaging implementation and the sub-grid calculation taking the emission heterogeneity into consideration may be of importance in terms of accurately calculating air pollutant abundance and their associated adverse health effects.

The aim of this study is to investigate segregation effects of heterogeneous emissions on O_3 levels in idealised urban street canyons, and to identify how segregation effects are influenced by the balance between chemistry and dynamics. The paper is structured as follows. In Section 2, the methodology based on photochemical box models is described in details, as well as the corresponding concept of intensity of segregation and the model scenarios. In the following sections, the results for prediction of ozone levels and the intensity of segregation are discussed.

2. Methodology

There are a large number of possible arrangements of street canyons in the urban canopy layer. In this study, we select two typical idealised urban street canyons as a representation. One large photochemical box model (hereafter referred to as the 'one-box' model) with averaged emissions of the two street canyons is used to represent the deterministic calculation based on the grid-average process; alternatively two small photochemical boxes (hereafter referred to as the 'two-box' model) are combined to represent two segregated street canyons with their own respective emissions. The photochemical box models (which assume that chemical species inside each box are well-mixed) can be simply applied and computationally inexpensive simulated. The model is written in FORTRAN77 language and run using FACSIMILE 4 integrator (Curtis and Sweetenham, 1987). A reduced chemical scheme (RCS), developed by Bright et al. (2013), is used as the chemical mechanism within the photochemical box models. The detailed model configuration is described as follows.

2.1. Model setup

Fig. 1 illustrates the overview of the box model configuration. It is assumed that in a cell of an urban air quality model, there are two street canyons with heterogeneous emissions represented by Box 1 and Box 2 with the same volume of air as indicated in the right panel (i.e. the 'two-box' model) of Fig. 1. There is no exchange between the two boxes, i.e. total segregation is assumed; we only consider exchange between the within-canyon air and the background air above the canopy layer. It is also assumed that the 'two-box' model represents the reality and the mean concentration,

$$C_{i,1+2} = (C_{i,1} + C_{i,2})/2 \quad (1)$$

represents the 'true' concentration of the i^{th} species in the canopy layer corresponding to this cell, with the concentrations in the 'one-box' model departing from this truth due to segregation effects. If a simplified approach of one single box (Box 0 indicated in the left panel of Fig. 1) is adopted in which the volume of Box 0 is the sum of the volumes of Box 1 and Box 2 (indicated in the right panel of Fig. 1) and $C_{i,0}$ is the modelled concentration from the 'one-box' model (Box 0 in Fig. 1), there would be an error for $C_{i,0}$ (either an overestimation or an underestimation) in comparison with the 'true' mean concentration $C_{i,1+2}$ derived from the 'two-box' model (Box 1 and Box 2 in Fig. 1). This error is expressed as

$$\Delta C_i = C_{i,0} - C_{i,1+2} \quad (2)$$