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Simulations of photochemical smog formation in complex urban areas

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HIGHLIGHTS

- An integrated CFD/CRD approach developed to simulate photochemical smog generation in complex urban areas.
- The Generic Reaction Set (GRS) model is proposed for chemical reactions.
- Shading effects within the street canyons are important for ozone production/depletion.
- Photochemical smog generation originating from traffic emission in city of Rotterdam successfully simulated.

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ABSTRACT

In the present study we numerically investigated the dispersion of photochemical reactive pollutants in complex urban areas by applying an integrated Computational Fluid Dynamics (CFD) and Computational Reaction Dynamics (CRD) approach. To model chemical reactions involved in smog generation, the Generic Reaction Set (GRS) approach is used. The GRS model was selected since it does not require detailed modeling of a large set of reactive components. Smog formation is modeled first in the case of an intensive traffic emission, subjected to low to moderate wind conditions in an idealized two-dimensional street canyon with a building aspect ratio (height/width) of one. It is found that Reactive Organic Components (ROC) play an important role in the chemistry of smog formation. In contrast to the NO_x/O_3 photochemical steady state model that predicts a depletion of the (ground level) ozone, the GRS model predicts generation of ozone. Secondly, the effect of direct sunlight and shadow within the street canyon on the chemical reaction dynamics is investigated for three characteristic solar angles (morning, midday and afternoon). Large differences of up to one order of magnitude are found in the ozone production for different solar angles. As a proof of concept for real urban areas, the integrated CFD/CRD approach is applied for a real scale (1×1 km^2) complex urban area (a district of the city of Rotterdam, The Netherlands) with high traffic emissions. The predicted pollutant concentration levels give realistic values that correspond to moderate to heavy smog. It is concluded that the integrated CFD/CRD method with the GRS model of chemical reactions is both accurate and numerically robust, and can be used for modeling of smog formation in complex urban areas.

1. Introduction

World-wide urbanization is already at a high level and is continuing to increase (an estimated 70% of the world's population will live in urban areas by 2030, Golden (2004)), with, correspondingly, a continuous degradation of urban air quality. In particular, the rise of mega-cities such as those in China and India has made the control of pollutant levels increasingly difficult. Human health is negatively impacted in several ways by such heavy air pollution. The study of air-borne pollutants, has, consequently, been conducted through direct measurements (e.g. Janssen et al. (1991), Kastner-Klein and Plate (1999), Tsai and Chen (2004), Boddy et al. (2005a, b), Clements et al. (2009)). as well as through numerical simulations (e.g. Garmory et al. (2009), Karim and Nolan (2011), Tominaga and Stathopoulos (2011), Wang et al. (2011),

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Gousseau et al. (2011), Kikumoto and Ooka (2012a, b), Tong et al. (2012), Yuan et al. (2014), Tong and Zhang (2015)). The simulations initially focused on passive pollutant dispersal in two-dimensional street canyons, without any active chemical reactions, as illustrated in Baik and Kim (1999). It has generally been found that pollutant dispersal is governed by vortex dynamics. For a building height-to-width (H/W) aspect-ratio of one, there is a single vortex that spans the whole canyon, while for a H/W ratio of two, two counter rotating vortices were found. The magnitude of pollutant concentrations is related to the size of the vortex, since this determines the air volume (assuming that concentration release is equal in both cases).

The research work of Baik and Kim (1999,2002) has lead to more modeling studies, where pollutant concentrations including different heating scenarios are studied (Kang et al., 2008; Tong and Leung, 2012). Reactive dispersion of NO_2, NO and O_3 has been studied (Baker et al. (2004), Kikumoto and Ooka (2012a)), where a Large Eddy Simulation (LES) framework is used. A background value for O_3 is assumed and NO_2 and NO are emitted at the lowest part of the urban canyon, resembling traffic emissions. A photo-stationary state defect was used to indicate whether the chemical reactions are near equilibrium. It was found to be a sensitive indicator of reactive mixing within the canyon, where this value is low in the center of the vortex.

The photo-stationary state defect was much higher at locations where ambient air was entrained into the canyon and close to the NO_2/NO emission source. The same chemical reactive pollutants were considered in a study by Kang et al. (2008), but that study also includes bottom heating of 5 K. The budget analysis of NO and NO_2 concentrations demonstrated that the magnitude of the advection or turbulent diffusion term was much larger than that of the chemical reaction term, confirming the importance of the proper modeling of turbulence parameters.

The chemical reaction studies discussed up to this point assumed reaction rates as a function of local air temperature, whereas in reality this is a photovoltaic process, i.e. determined by sunlight intensity. Using only air temperature as a proxy for photovoltaic processes incorrectly predicts chemical reactions during the night, or in the shadow of buildings. Grawe et al. (2007) performed LES simulations to study reactive pollutants including shadowing effects by setting the reaction rate to 25% in the shaded regions. It was shown that the effect of local shading has a significant impact on the magnitude of the pollutant concentration. The geometrical layout of the shading within the canyon and the wind speed in the canyon was shown to affect the spatial distribution of the pollutants, rather than their overall magnitude. The numerical studies discussed up to now have all used the NO_x/O_3 photochemical steady state model, neglecting the influence of Reactive Organic Components (ROC), Carpenter et al. (1998). Thus, the system under study consists of three species (O_3, NO_2, NO) and three reactions which connect them. The more complex Carbon Bond IV (CB-IV) model was studied by Kwak and Baik (2012) and Kwak et al. (2013). However, this model is computationally very demanding in that it comprises some 20 chemical species, 70 reactions, and a set of 28 PDEs which must be solved. A less demanding model, the Generic Reaction Set (GRS) mechanism, is also available. It has been studied against CB-IV model in photochemical smog-chamber studies by Azzi and Johnson (1992), and was found to compare well using a set of just 7 categories of (grouped) chemical species, including ROCs. The GRS mechanism has been successfully applied in several studies, at scales as large as several hundreds of kilometers (Venkatram et al. (1997), Abdul-Wahab et al. (2002), Lagzi et al. (2008), Pournazeri et al. (2014)). In the present study the GRS is used for computation of smog formation at the urban scale, including ROC's. The goal of this paper is to gain more insights into the dynamics of photochemical smog formation within and above complex urban areas at a neighborhood scale (few kilometers) using the integrated CFD/CRD approach with the GRS model of chemical reactions involved in smog generation.

2. Governing equations

2.1. Computational Fluid Dynamics

By assuming incompressible and steady flow conditions, the long-term time-averaged conservation of mass and momentum can be written as

$$\frac{\partial U_i}{\partial x_i} = 0 \tag{1}$$

$$U_j \frac{\partial U_i}{\partial x_j} = -\frac{1}{\rho}\frac{\partial P}{\partial x_i} + \frac{\partial}{\partial x_j}\left[\nu\left(\frac{\partial U_i}{\partial x_j} + \frac{\partial U_j}{\partial x_i}\right) - \overline{u'_i u'_j}\right] \tag{2}$$

Here, U_i is the mean velocity, P is the pressure, ρ the density and ν the kinematic viscosity. The first-order closure in the form of the two-equation $k-\varepsilon$ model is used in this study. The Reynolds stress tensor $\overline{u'_i u'_j}$ is modeled using the Boussinesq approximation as:

$$\overline{u'_i u'_j} = \frac{2}{3}k\delta_{ij} - \nu_t\left(\frac{\partial U_i}{\partial x_j} + \frac{\partial U_j}{\partial x_i}\right) \tag{3}$$

The transport equation for the turbulent kinetic energy (k) and its dissipation rate (ε) are:

$$U_j \frac{\partial k}{\partial x_j} = \frac{\partial}{\partial x_j}\left[\left(\nu + \frac{\nu_t}{\sigma_k}\right)\frac{\partial k}{\partial x_j}\right] + P_k - \varepsilon \tag{4}$$

$$U_j \frac{\partial \varepsilon}{\partial x_j} = \frac{\partial}{\partial x_j}\left[\left(\nu + \frac{\nu_t}{\sigma_\varepsilon}\right)\frac{\partial \varepsilon}{\partial x_j}\right] + C_{\varepsilon 1}\frac{\varepsilon}{k}P_k - C_{\varepsilon 2}\frac{\varepsilon^2}{k} \tag{5}$$

where the production of turbulent kinetic energy (P_k) is modeled as

$$P_k = 2\nu_t S_{ij} S_{ji} \tag{6}$$

and $S_{ij} = \frac{1}{2}\left(\frac{\partial U_i}{\partial x_j} + \frac{\partial U_j}{\partial x_i}\right)$ is the mean rate of strain tensor. The turbulent viscosity (ν_t) is calculated as

$$\nu_t = C_\mu k\tau \tag{7}$$

with τ as a characteristic turbulent time scale. In the present study we applied the time-scale limiter, proposed by Durbin (1996), in order to reduce the over-prediction of the turbulent kinetic energy in stagnant regions or regions with a strong curvature of the flow. The application of Durbin limiter significantly improved flow predictions in configurations mimicking complex urban areas, as previously shown in Hanjalić and Kenjereš (2008), Kenjereš and ter Kuile (2013) and Kenjereš et al. (2015). The turbulent time scale (τ) is calculated as:

$$\tau = \min\left[\frac{k}{\varepsilon}, \frac{0.6}{\sqrt{6}C_\mu|S|}\right] \tag{8}$$

where $|S|$ is the square root of the modulus of the mean rate of strain tensor. The standard coefficients in the $k-\varepsilon$ model are used: $C_\mu = 0.09$, $\sigma_k = 1.0$, $\sigma_\varepsilon = 1.3$, $C_{\varepsilon 1} = 1.44$, and $C_{\varepsilon 2} = 1.92$.