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Modelling traffic-induced multicomponent ultrafine particles in urban street canyon compartments: Factors that inhibit mixing☆

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ABSTRACT

This study implements a two-box model coupled with ultrafine particle (UFP) multicomponent microphysics for a compartmentalised street canyon. Canyon compartmentalisation can be described parsimoniously by three parameters relating to the features of the canyon and the atmospheric state outside the canyon, i.e. the heterogeneity coefficient, the vortex-to-vortex exchange velocity, and the box height ratio. The quasi-steady solutions for the two compartments represent a balance among emissions, microphysical aerosol dynamics (i.e. evaporation/condensation of semi-volatiles, SVOCs), and exchange processes, none of which is negligible. This coupled two-box model can capture significant contrasts in UFP number concentrations and a measure of the volatility of the multi-SVOC-particles in the lower and upper canyon. Modelled ground-level UFP number concentrations vary across nucleation, Aitken, and accumulation particle modes as well-defined monotonic functions of canyon compartmentalisation parameters. Compared with the two-box model, a classic one-box model (without canyon compartmentalisation) leads to underestimation of UFP number concentrations by several tens of percent typically. By quantifying the effects of canyon compartmentalisation, this study provides a framework for understanding how canyon geometry and the presence of street trees, street furniture, and architectural features interact with the large-scale atmospheric flow to determine ground-level pollutant concentrations.

1. Introduction

Urban air pollution induced by road traffic is a key environmental concern (Murena et al., 2009). As one of the major urban pollutants, particulate matter (PM) has received much attention in the scientific community (Dall'Osto et al., 2011; Heal et al., 2012). PM_{10} (with an aerodynamic diameter $d_p < 10\,\mu m$) and $PM_{2.5}$ ($d_p < 2.5\,\mu m$) are currently regulated in terms of the mass concentrations of particles (US EPA, 2017b; European Commission, 2017). Although regulations for ultrafine particle (UFP or $PM_{0.1}$, $d_p < 0.1\,\mu m$) do not yet exist, UFP is a very significant contribution to total particle number concentrations (Harrison et al., 2000). UFP may accumulate in the lungs (Panis et al., 2010) or penetrate cells/tissue (Geiser et al., 2005), causing health effects because of their small sizes. Semi-volatile components of UFP may also contribute to secondary organic aerosol formation (Baldauf et al., 2016).

An urban street canyon is a linear urban feature having buildings on both sides of a street (Li et al., 2008). In such an environment, ground-level atmospheric flow is restricted by the buildings, which may lead to reduced air ventilation between the street canyon and the overlying atmospheric background (Salim et al., 2011). According to the canyon aspect ratio (*AR*, the ratio of building height *H* to street width *W*), street canyons may be categorized into deep ($AR \geq 2$), regular ($0 < AR < 2$), and avenue ($AR \leq 0.5$) (Vardoulakis et al., 2003). Deep street canyons present worst-case scenarios for the dispersion of air pollutants (Li et al., 2009), since there may be multiple segregated vortices formed in the canyon, which can lead to even poorer ventilation conditions. Below, we call such segregated inhibition of mixing within the street canyon, *compartmentalisation*. The presence of street trees, street furniture, and architectural features can also lead to compartmentalisation in

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shallower street canyons and may create multiple split vortices with reduced exchange. Street trees (Gromke et al., 2008) or architectural elements, e.g. roof shapes (Takano and Moonen, 2013), balconies (Murena and Mele, 2016) and elevated expressways (Huang and Zhou, 2013), may produce an internal 'lid' that constrains the height of the primary street vortex (Gromke and Ruck, 2007).

The microphysical and/or chemical processes associated with mixing across compartments, together with emissions and the exchange with background air can be parsimoniously represented by a two-box model. The concept of a two-box model was previously introduced and evaluated against field measurements (Murena et al., 2011; Murena, 2012) to predict carbon monoxide (CO) concentration (taken as a passive scalar because of its long chemical lifetime) in a deep street canyon and no chemical processes were considered. The traditional one-box model (originally assuming a single vortex in a regular canyon) may not be appropriate for deep street canyon scenarios (with canyon compartmentalisation) (Murena et al., 2011; Murena, 2012). Zhong et al. (2015) adopted simple NO_x-O_3 (nitrogen oxides-ozone) photochemistry into a two-box model (representing two segregated vortices found in their large eddy simulation LES of a deep canyon with AR = 2) and there was a good agreement between the LES model and the two-box model. Zhong et al. (2017) further coupled more complex O_3-NO_x-VOC (nitrogen oxides-ozone-volatile organic compounds) chemistry into both LES and a two-box model for a deep street canyon. Concentrations of oxidants were found to be increased by about 30–40% via the additional OH/HO_2 (hydroxyl/hydroperoxyl radicals) chemistry compared with simple NO_x-O_3 photochemistry adopted in Zhong et al. (2015). The pre-processing within the canyon could enhance oxidant fluxes from the canyon to the overlying atmospheric background, with an even greater effect for deep street canyons than shallower street canyons. Zhong et al. (2016) employed the two-box model coupled with O_3-NO_x-VOC chemistry to investigate effects of governing parameters (i.e. heterogeneity coefficient, exchange velocity and box height ratio) for a variety of emission scenarios and to identify under which conditions NO_2 (nitrogen dioxide) at the pedestrian level would exceed its air quality limit value.

The current study extends the two-box modelling approach by including the multicomponent microphysics of UFP in urban street canyon compartments. The canyon-box modelling approach is similar conceptually to that of Pugh et al. (2012b) but has been coded independently. The UFP code for the present study is shared with that of CiTTy-Street-UFP (Nikolova et al., 2016), i.e. the CiTTyCAT (Pugh et al., 2012a) model coupled with UFP microphysics.

2. Methods

2.1. Framework of a two-box model coupled with UFP

The two-box model based on vortex structure from the LES model for a deep street canyon (AR = 2) was previously implemented for both simple NO_x-O_3 and more complex O_3-NO_x-VOC chemistry, and evaluated against the LES-chemistry models (Zhong et al., 2015, 2016, 2017). The extension of this simplified two-box model to the multicomponent microphysics of UFP concerning emissions, microphysical aerosol dynamics (i.e. evaporation/condensation of semi-volatiles, SVOCs), and exchange processes in a compartmentalised street canyon (Fig. 1) for both particulate and gas phases is described below. For the particulate phase:

$$\frac{dQ_{q,j,U}}{dt} = \frac{w_{t,L}}{H_U}\left(N_{j,L} - N_{j,U}\right)\chi_{q,j,L}m_j - \frac{w_{t,U}}{H_U}\left(N_{j,U} - N_{j,b}\right)\chi_{q,j,U}m_j + \Delta Q_{q,j,U} \quad (1)$$

$$\frac{dQ_{q,j,L}}{dt} = -\frac{w_{t,L}}{H_L}\left(N_{j,L} - N_{j,U}\right)\chi_{q,j,L}m_j + E_{q,j,L} + \Delta Q_{q,j,L} \quad (2)$$

where "$_q$" represents the component q; "j" is the size bin j; "L" and "U" represent the lower and upper boxes, respectively; "$_b$" represents the overlying background; "Q" denotes the mass concentration in the particulate phase; "N" is the number concentration; "χ" is the mass fraction; "m" is the mass of one representative particle in a sectional bin; "w_t" is the exchange velocity (the exchange/diffusion process are based on the number concentration gradient); "H" is the height of the box; "E" is the emission rate into the lower box volume per unit time; "ΔQ denotes the source terms for the particulate phase from the UFP module due to aerosol transformation processes (e.g. condensation/evaporation in this study).

For the gas phase,

$$\frac{dc_{q,U}}{dt} = \frac{w_{t,L}}{H_U}\left(c_{q,L} - c_{i,U}\right) - \frac{w_{t,U}}{H_U}\left(c_{q,U} - c_{q,b}\right) + \Delta c_{q,U} \quad (3)$$

$$\frac{dc_{q,L}}{dt} = -\frac{w_{t,L}}{H_L}\left(c_{q,L} - c_{q,U}\right) + E_{q,L} + \Delta c_{q,L} \quad (4)$$

where c is the mass concentration in the gas phase; Δc denotes the source terms for the gas phase from the UFP module due to aerosol transformation processes; other symbols are same as those in Equations (1) and (2). In this study, the source terms (Equations (1)–(4)) are derived from the UFP module due to particle condensation/evaporation (further details in Section 2.2), rather than from the chemistry module in previous studies (Zhong et al., 2015, 2016, 2017). The number of UFP components used in the model is 18: 1 non-volatile core and 17 surrogate Semi-Volatile Organic Compounds (SVOC) (parameterised as n-alkanes from $C_{16}H_{34}$ to $C_{32}H_{66}$) (Nikolova et al., 2016). The present model runs use 15 sectional size bins, ranging from 6.7 nm to 501.4 nm in a uniform logarithmic scale. The UFP number concentration in a size bin is calculated based on the total mass concentrations in a size bin (divided by the dry aerosol mass per particle in the given size bin). There are 17 tracers in the gas-phase corresponding to each SVOC component. Sequential ordinary differential equations in the model are solved on a 0.3 s time step for emission/exchange processes and adaptive time steps for aerosol evaporation/condensation processes.

For deep canyons (AR ≥ 2), the spontaneous formation of primary and secondary vortices motivates the use of multiple boxes (Fig. 1a and b); for other values of AR, street trees, street furniture, and architectural features may all lead to zones of inhibited mixing (Fig. 1c and d) that motivate a multi-box approach (Gromke and Ruck, 2007; Huang and Zhou, 2013; Gromke et al., 2008).

2.2. Condensation/evaporation of semi-volatiles

The condensation/evaporation process of semi-volatiles (SVOCs) is one of the most important aerosol transformation processes in predicting the fate of ultrafine particles in urban air (Harrison et al., 2016). This process is driven by the difference between the partial pressure of a gas species and its saturation vapour pressure over a particle surface (Jacobson, 2005), which will alter the size of the particle. The condensation/evaporation rate of each component (q)