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The impact of urban canopy meteorological forcing on summer photochemistry

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ABSTRACT

The regional climate model RegCM4.4, including the surface model CLM4.5, was offline coupled to the chemistry transport model CAMx version 6.30 in order to investigate the impact of the urban canopy induced meteorological changes on the longterm summer photochemistry over central Europe for the 2001–2005 period. First, the urban canopy impact on the meteorological conditions was calculated performing a reference experiment without urban landsurface considered and an experiment with urban surfaces modeled with the urban parameterization within the CLM4.5 model. In accordance with expectations, strong increases of urban surface temperatures (up to 2–3 K), decreases of wind speed (up to -1 ms^{-1}) and increases of vertical turbulent diffusion coefficient (up to 60–70 m^{2}s^{-1}) were found. For the impact on chemistry, these three components were considered. Additionally, we accounted for the effect of temperature enhanced biogenic emission increase. Several experiments were performed by adding these effects one-by-one to the total impact: i.e., first, only the urban temperature impact was considered driving the chemistry model; secondly, the wind impact was added and so on.

We found that the impact on biogenic emission account for minor changes in the concentrations of ozone (O_3), oxides of nitrogen NOx = NO + NO_2 and nitric acid (HNO_3). On the other hand, the dominating component acting is the increased vertical mixing, resulting in up to 5 ppbv increase of urban ozone concentrations while causing -2 to -3 ppbv decreases and around 1 ppbv increases of NOx and HNO_3 surface concentrations, respectively. The temperature impact alone results in reduction of ozone, increase in NO, decrease in NO_2 and increases of HNO_3. The wind impact leads, over urban areas, to ozone decreases, increases of NOx and a slight increase in HNO_3. The overall impact is similar to the impact of increased vertical mixing alone. The Process Analysis (PA) technique implemented in CAMx was adopted to investigate the causes of the modeled impacts in more details. It showed that the main process contributing to the temperature impact on ozone is a dry-deposition enhancement, while the dominating process controlling the wind impact on ozone over cities is the advection reduction. In case of the impact of enhanced turbulence, PA suggests that ozone increases are, again as assumed, the result of increased downward vertical mixing supported by reduced chemical loss. Comparing the model concentrations with measurements over urban areas, a slight improvement of the model performance was achieved during afternoon hours if urban canopy forcing on chemistry via meteorology was accounted for. The study demonstrates that disregarding the urban canopy induced meteorological effects in air-quality oriented modeling studies can lead to erroneous results in the calculated species concentrations. However, it also shows that the individual components are not equally important: urban canopy induced turbulence effects dominate while the wind-speed and temperature related ones are of considerably smaller magnitude.

1. Introduction

Cities and urban areas impact the environment in many ways, however, an impact on the atmospheric environment is probably the 'most important and most far-reaching' (Folberth et al., 2015). Both meteorology and atmospheric chemistry are influenced: (i) urban areas are largely covered by artificial surfaces and they affect mechanical and thermodynamical properties of the air above in a very specific way leading to higher temperatures, lower winds, increased turbulence etc. (Lee et al., 2011; Huszar et al., 2014). (ii) Cities further represent strong emission centres releasing large quantities of gaseous material and aerosol into atmosphere directly influencing air-quality and atmospheric chemistry in general (Timothy and Lawrence, 2009; Huszar et al., 2016a). (iii) These emissions then may lead to modification of

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radiative and thermal balance, cloud properties and climate (Seinfeld and Pandis, 1998; Folberth et al., 2015; Huszár et al., 2016b). There is further a secondary pathway how air chemistry is influenced: (iv) urban canopy induced meteorological changes directly link to changes in air chemistry influencing chemical reactions, transport, turbulence, deposition and emission.

Generation of the so called urban heat island (UHI) and, in general, the complex nature of the influence of the urban-canopy on meteorological conditions and climate have been identified since the early 80s (Oke, 1982). Since then, UHI and related meteorological effects were subject to large number of studies investigating the impact on temperature (Basara et al., 2008; Gaffin et al., 2008); humidity (Richards, 2004); turbulence and wind-speed (Roth, 2000; Kastner-Klein et al., 2001; Hou et al., 2013); structure of the boundary layer (Angevine et al., 2003); precipitation and hydrological cycle (Rozoff et al., 2003). With the introduction urban schemes, starting from very simple land-use parameters modification up to complex multilayer urban-canopy parameterizations/models, modeling approaches to urban effects on meteorology and climate became widespread. These examined both local (Flagg and Taylor, 2011; Wouters et al., 2013; Hou et al., 2013) and regionals scales (Feng et al., 2013; Trusilova et al., 2008; Struzewska and Kaminski, 2012; Huszar et al., 2014). It was further shown that cities' impact on meteorology and climate is usually not limited to the geographical location of the city itself, but the impact propagates to regional scales and that urbanization can contribute even to regional warming (e.g. Huszar et al., 2014).

These urban canopy induced meteorological perturbations, as already pointed out, must have impact on air-quality, as atmospheric chemistry is largely controlled by meteorological conditions. Elevated urban temperatures in connection with UHI increase reaction rates resulting in faster chemical conversions. Higher temperatures further modify dry and wet scavenging (Seinfeld and Pandis, 1998) and influence wind by triggering urban-breeze circulation which can help the circulation of pollutants from as well as to urban areas depending on the daytime (Ryu et al., 2013a; Hidalgo et al., 2010). On the other hand, drag induced wind stilling over urban areas can slow down pollution dispersion into larger scales, for both primary pollutants and secondary formed ones. Increased turbulence over urban areas and stronger eddy-diffusion supports vertical transport of pollutants away from the urban canopy layer often leading to high gradients of primary and secondary pollutants resulting in elevated concentrations in upper layers of the planetary boundary layer (PBL; Stutz et al., 2004). At last, urban areas are almost always to some degree covered by vegetation (parks, urban green belts etc.) and the biogenic emissions released are enhanced by the temperature increase due to UHI (Song et al., 2008).

To evaluate an integrated effect of the urban induced meteorological changes listed above, several difficulties appear. First of all, the individual impacts are often counteracting. For example elevated temperatures trigger higher reaction rates for reactions that are responsible for both ozone production and destruction at the same time. Slower winds keep primary pollutants closer to sources where they trigger formation of secondary pollutants, however, at the same time can contribute to their more effective destruction, like in case of NOx-ozone interaction (formation vs. titration). Enhanced vertical turbulence removes primary pollutants like NOx from urban areas, which can suppress ozone titration leading to increase of concentrations. Simultaneously however, the secondary produced pollutants are subject to vertical eddy-removal which, consequently, lowers their concentrations.

Secondly, the urban induced meteorological changes are not uniform in time and have a very specific daily cycle (see further). This holds for the emissions as well. Consequently, as a result of different timing of positive and negative peaks in both, additional complexity in urban air-quality/meteorology interactions is introduced. It becomes clear that the use of integrated modeling tools for both meteorology and air-chemistry is a necessity.

There have been many model based efforts to analyze the urban triggered meteorological changes and the forcing they represent on chemistry. Martilli et al. (2003) using a mesoscale model offline coupled to a chemistry transport model analyzed the impact of using urban canopy parameterization on meteorological conditions and subsequently on NOx and ozone concentrations. They found that considering urban effects in mesoscale meteorology-chemistry simulation clearly reduces model biases. However, they focused on a single city, Athens (Greece) and simulated only one day. Civerolo et al. (2007), using a bulk (urban landuse treated as any other landuse type with appropriately modified properties) approach for urban surfaces, investigated the impact of future urbanization on ozone and found significant increase, especially in terms of the episode-maximum 8-h average. Sarrat et al. (2006) studied Paris region ozone-NOx photochemistry modifications due to urban canopy forcing. They found that the main driver for chemistry changes is an enhancement of turbulence over urban areas. Wang et al. (2007) predicts increase in O_3 concentration, resulted from nocturnal urban heat island effects and wind speed reduction over the highly urbanized Perl River Delta region in China. Wang et al. (2009) arrived at a similar conclusion. A large effort was put to analyze the urbanization impact on air quality over China in many other studies (e.g. Xie et al., 2016; Zhu et al., 2017, and references therein) but they simulated only one episode of a few days length up to one month. An important finding was presented by Jiang et al. (2008), who, using WRF coupled with a single layer urban canopy model, looked at the individual as well as combined impact of urbanization, climate change and emission changes on ozone levels. They predicted concentration increase due to urbanization of a similar magnitude compared to changes due to other two influences strongly justifying considering the urban canopy effects on chemistry in modeling studies. The area of Seoul, South Korea was examined in high resolution by Ryu et al. (2013a) and significant changes in ozone concentrations were found, caused by the urban-breeze circulation effect already mentioned, modulated further by the anthropogenic heat (AH) enhanced PBL height and modified mixing (Ryu et al., 2013b). Recently, Fallmann et al. (2016) investigated an impact of urban greening utilization of green roofs on air quality and found improvement in O_3 due to lower mitigated temperatures increase, however, due to decreased mixing, some primary pollutants had higher concentrations.

There is a large number of studies that investigated urban boundary layer chemistry. Especially the nocturnal urban PBL chemistry has received great attention (e.g. Benton et al., 2010) as well as measurements and modeling of the vertical profiles in PBL (Stutz et al., 2004; Geyer and Stutz, 2004; Brown et al., 2006; Benton et al., 2010). These and many others (see references inside) claim the complex non-linear nature of the interaction between urban PBL dynamics and air composition justifying models as an inevitable tool.

This work contributes to the listed modeling studies introducing three novelties: 1) it analyzes the long-term impact of urban canopy effects on air quality considering multiple years (2001–2005), 2) it covers a regional domain where a large number of medium to big cities is located, enabling to perform a more robust multi-city analysis instead of focusing on a particular urban area (as is the case in many of the listed studies), 3) besides the combined impact this work concerns on the impacts of individual components as well. This opens the door to a more detailed analysis of which processes are encompassed in the overall impact and to what degree they contribute to it.

2. Models and experimental design

2.1. Models

2.1.1. Regional climate model

As a meteorological driver, regional climate model RegCM4 (version 4.4) was applied (Giorgi et al., 2012). For large-scale precipitation, the SUBEX scheme (Pal et al., 2000), for convection the Grell scheme