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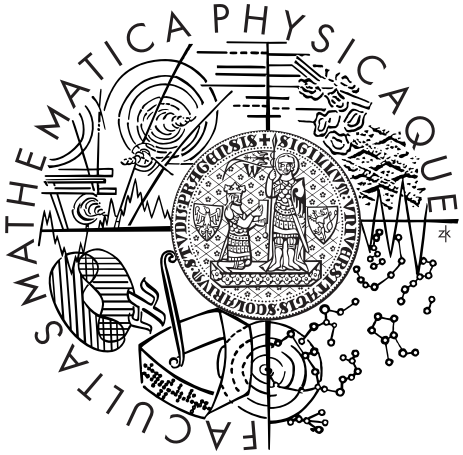

Peter Huszár

Impact of urbanization on climate and atmospheric chemistry

Prague 2021

To my beloved wife

Contents

Preface

The presented thesis entitled *Impact of urbanization on climate and atmospheric chemistry* summarizes recent developments in the research of environmental imprint of urbanization on atmosphere. Cities and urban areas in general influence the environment in many ways, while the most pronounced and far reaching is the impact on the atmosphere. This follows multiple direct and indirect pathways. Firstly, urban areas are largely covered by artificial surfaces and are clearly distinguished from natural ones by mechanical, radiative, thermal and hydraulic properties substantially altering the fluxes of energy, momentum and moisture. This directly impacts the meteorological conditions. Secondly, cities emit large amount of gaseous material and aerosol into the air directly influencing air-quality and atmospheric chemistry in general. Thirdly, part of the emitted material interacts with solar and thermal radiation altering thus the radiative and thermal balance of the atmosphere, which in turn impacts the climate. At last, the meteorological changes forced by the artificial surfaces typical for urban areas further influence atmospheric chemistry by modulating the formation, dispersion and deposition of pollutants.

These effects are strongest on local scale, i.e. close to emission sources within city outskirts. On the other hand, on global scale impacts are detectable as well but are much smaller. Here, we focused on regional scale effects and interactions, where they can vary substantially depending on the urban density. Given the complexity of interactions between emissions, atmospheric chemistry and climate, which includes many feedbacks, regional scale numerical modelling approaches to both meteorology and atmospheric chemistry are crucial to be adopted.

Accordingly, we applied a regional climate model coupled to chemistry transport model to investigate each of the above mentioned chain of effects including some of such effects. We focused on the region of central Europe which is characterized by medium urban density. The results were published by Peter Huszár in eight peer-reviewed papers: Huszar et al. (2014), Huszar et al. (2016a), Huszar et al. (2016b), Huszar et al. (2018a), Huszar et et. (2018b), Huszar et al. (2020a), Huszar et al. (2020b) and, finally, Huszar et al. (2021).

First the impact of urban canopy on meteorological conditions and climate was evaluated for which we had to extend a regional climate model with an urban canopy sub-model that accounts for the specific physical properties of urban surfaces and objects. The results were published in Huszar et al. (2014) titled *Regional climate model assessment of the urban land-surface forcing over central Europe*. Next, in Huszar et al. (2016b) entitled *On the long-term impact of emissions from central European cities on regional air quality*, we turned our attention on the second direct impact of urban areas on environment, i.e. the impact of urban emissions on regional air-quality, where we were specifically interested at how city emissions contribute to both urban and rural concentrations of pollutants and how far the impact reaches. After qualifying and quantifying the above mentioned direct influences via urban canopy meteorological effects and emissions, we evaluated the secondary impacts acting trough radiative effects of primary emissions and secondary pollutants and trough the urban canopy meteorological forcing on tropospheric chemistry. The study, titled *The regional impact of urban emissions on climate over central Europe: present and future emission perspectives* published in Huszar et al.(2016b) arrives to important conclusions about the regional climate impacts of urban emissions indicating rather small temperature effects. In two studies titled *The impact of urban canopy meteorological forcing on summer photochemistry* and *Impact of urban canopy meteorological forcing on aerosol concentrations*

(Huszar et al., 2018a,b) we investigated a further pathway of climate/air-quality interactions over urbanized areas: how is the chemistry and transport of pollutants, both gas-phase and particulate ones, altered by the urban canopy meteorological effects; in this study we further introduced a new urban canopy model in the regional climate model adopted. In these too latest papers it was revealed that turbulence plays a crucial role in urbanization induced modifications of air-quality. Motivated by this, we set-up a series of model experiments in the study titled *Urban canopy meteorological forcing and its impact on ozone and PM2.5: role of vertical turbulent transport* (Huszar et al., 2020a) were we focused on the urban induced modification of turbulent fluxes and their implication on pollutant concentrations. This later study also introduced the term "urban canopy meteorological forcing" (UCMF) which better reflects the physical concept behind the studied processes. In the paper called *The impact of urban land-surface on extreme air pollution over central Europe* (Huszar et al., 2020b) the attention turned to the UCMF impact on the high end of the pollutant concentration distribution as extreme air pollution is of much greater policy interest. Finally, based on the last four studies, it was postulated that UCMF will probably influence the regional footprint of urban emissions (how they modify the background air chemistry) and this assumption was confirmed in our most recent study titled *The regional impact of urban emissions on air quality in Europe: the role of the urban canopy effects* (Huszar et al., 2021).

The research performed within the presented studies was greatly motivated by the fact, that already more than half of the human population lives in cities while urbanization is still an ongoing process. To ensure the quality of life of their inhabitants, it is of great importance to study how cities interact with the atmospheric environment.

Peter Huszár

Abstract

Cities cover only a few percent of Earth's surface with, however, more the half of the population inhabiting them. This makes them hotspots of human activities therefor strong sources of concentrated emissions. Moreover, cities are covered with surfaces substantially different from natural ones which generates additional forcing on atmosphere greatly modifying the meteorological conditions and climate. Due to large urban population there is a high demand to investigate how these effects act in detail and to qualify and quantify the role of interactions between urban areas, air-chemistry and climate.

Using are regional climate models coupled to chemistry transport models, we investigate these interactions over Europe for present day conditions. The following pathways of urban influence on atmospheric environment are considered: the direct impact of urban surfaces on meteorological conditions and climate, the direct impact of urban emission on tropospheric chemistry, the indirect impact of urban emission caused chemistry perturbations on atmospheric radiation and climate and, lastly, the impact of urban canopy induced meteorological forcing on tropospheric chemistry.

The impact of urban surfaces on climate is characterized by a clear increase of average temperatures by up to 1-2 K over cities as summer average. The impact on wind speed is characterized by decreases up to 1 ms^{-1}, peaking over cities. We further found that the additional drag introduced by urban surfaces increase the vertical turbulent diffusion by 60–70 m^2s^{-1} over cities and is almost uniform throughout the day. This increase is closely connected to the increased boundary layer height, which is elevated by more than 100 meters due to urban surfaces, especially during summer. Urban surfaces decrease water vapor mixing ratio by up to 1 $g.kg^{-1}$ due to low evaporation and high run-off. Low evaporation is also responsible for suppressed precipitation over cities by up to few millimeters per day.

A strong impact on regional scale air quality due to urban emission was simulated too. Significant ozone titration occurs over cities while over rural areas remote from cities, ozone production is modeled, mainly in terms of number of exceedances and accumulated exceedances over the threshold of 40 ppbv. It is further found that urban nitrogen oxides (NO_x), sulfur dioxide (SO_2) and fine aerosol particle ($PM_{2.5}$) emissions also significantly contribute to concentrations in the cities themselves (up to 50–70 % for NO_x and SO_2, and up to 60 % for $PM_{2.5}$), but the contribution is large over rural areas as well (10–20 %). Although air pollution over cities is largely determined by the local urban emissions, a considerable (often a few tens of %) fraction of the concentration is attributable to other sources from rural areas and minor cities. For the case of Prague, it was further shown that the inter-urban interference between large cities does not play an important role which means that the impact on a chosen city of emissions from all other large cities is minor. At last, it is shown that to achieve significant ozone reduction over cities in central Europe, the emission control strategies have to focus on the reduction of nonmethane volatile organic compounds (NMVOC), as reducing NO_x (due to suppressed titration) leads often to increased O_3 surface levels. The influence over rural areas is however always in favor of improved air-quality, i.e. both NO_x and/or NMVOC reduction ends up in decreased ozone pollution, mainly in terms of exceedances.

As expected, the simulated changes in species concentrations may result in the perturbation of radiative balance if these species have the ability to interact with radiation. Indeed, this is the case for ozone and for aerosol and statistically significant impact on climate is modeled. It is characterized by a statistically significant cooling up to -0.02 K

and -0.04 K in winter (DJF) and summer (JJA) season, respectively, mainly over cities. We found that the main contributor to the cooling is the aerosols direct and indirect effects, while the ozone titration, calculated especially for DJF, plays rather a minor role. In accordance with the vertical extent of the urban emission induced aerosol perturbation, cooling dominates the first couple of model layers up a few hundreds meters. We also found a clear diurnal cycle of the radiative impacts with maximum cooling just after noon (JJA) or later in afternoon (DJF). Furthermore, statistically significant decreases of surface solar radiation are modeled, in accordance with the temperature decrease. The impact on the boundary layer height is small but statistically significant and reaches 1 m and 6 m decreases in DJF and JJA, respectively. We did not find any statistically significant impact on precipitation and wind speed. Comparing the climate impact of emissions to the impact due to urban canopy forcing, it is clear that the urban emission induced climate effects are of rather minor importance, at least when investigating the non-CO_2 effects.

As air-quality and atmospheric chemistry in general is closely connected to the meteorological conditions, it is clear that the simulated urban canopy induced changes in meteorology/climate (the so called urban canopy meteorological forcing – UCMF) will have implications in the concentration of pollutants. Indeed, we simulate statistically significant changes in NOx, nitric acid (HNO_3) and ozone concentrations over cities due to urban canopy meteorological effects, more specifically due to temperature-, wind- and turbulence modifications. 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. It was further shown, that turbulence is the main process that shapes the overall UCMF impact on aerosol concentrations: $PM_{2.5}$ is reduced by -3 μgm^{-3} if UCMF is considered, while the impact of temperature modifications is a decrease too (due to weaker gas-to-particle formation), counteracting the increase caused by decreased urban wind speeds. It was also shown, that turbulence is the primary component of the UCMF regardless of the chosen parameterization for PBL or, in particular, for the vertical eddy diffusivity.

When focusing on extreme air pollution, the UCMF related air–quality impacts are larger not only in absolute sense, but also in relative numbers. This holds strongly for NO_2 and $PM2.5$, while in case of ozone, the impact on average values is of comparable magnitude than that on the high end of the distribution. Finally, if UCMF is accounted for in the calculation of the local and regional footprint of urban emissions, their impact is smaller. In case of NO_2 and $PM2.5$ it is a 40 to 60% decrease. For ozone, for which urban emissions cause decrease over urban areas. This decrease is about 60% smaller in magnitude if UCMF is considered.

1. Introduction

Anthropogenic emissions are not distributed uniformly over the globe. Similarly, the population density varies from region to region in orders of magnitude. This creates a potential for uneven distribution of the impacts of these emissions on environment and on human life. With higher population density higher density of human activities appears and this results in more concentrated emissions and impacts at the same time.

The socio-economic circumstances often force human to live in cities. They represent these spots of high population density. Year 2009 was the first reported year with more than 50% of the world's population living in cities (UN, 2009), occupying less than 0.1 % of the Earth's surface. According to the United Nation's report (UN, 2008), in Europe in 2008, 73% of the population inhabit cities and by the mid 21th century, this is predicted to reach 84 %, representing a rise from 531 to 582 million in absolute numbers. For Czech Republic, a similar change, from 73.5% to 83% is projected by the Czech Statistical Office.

Consequently, the investigation of urban environmental conditions and factors that influence them is becoming extremely important. This means to evaluate how cities influence their own environment and, on the other hand, how their surroundings impact the environment in them. Among the many types of urban impact on environment, Folberth et al. (2015) defines the impact on atmospheric environment as the 'most important and most far-reaching' while atmospheric environment here means the combination of meteorological conditions and air chemistry.

Urban areas influence the atmosphere directly and indirectly. There are two ways of the direct impact: (i) by emission of pollutants altering the air composition and (ii) via meteorological forcing caused by artificial surfaces characteristic for urban areas. Indirect pathways of urban influence of atmosphere include: iii) modifying the radiative and thermal balance by changing of radiatively active gas and aerosol concentrations by urban emissions (consequence of (i)), and iv) modifying the chemical processes and transport of pollutants due to modifications of urban meteorology (consequence of (iii)).

In the next section, individual pathways are described including the general interactions (i.e. not only over cities) between atmospheric chemistry and climate. This will be followed, in Chapters 2 to 9, by the regional scale climate-chemistry model assessment of these interactions for present day conditions over Europe.

1.1 Direct impact of urban areas on atmosphere

1.2 Urban emissions

Due to energy production (e.g. heating), transportation and industrial activity concentrated over small area, urbanization results in increasing air pollution emissions. These emissions cause significant burden of different species in atmosphere, resulting in modified chemical composition and chemistry of the air. Two families of species released from cities can be distinguished. First family are comprised by the long-lived green-house-gases (GHG; carbon dioxide, methane and nitrous oxide). They absorb and re-emit the thermal radiation coming from the Earth and thus heating it. Cities with population larger than 10 million (Megacities – often by definition) are found to contribute with around 12 % to the anthropogenic emissions of carbon dioxide and lesser fractions to methane and nitrous oxide (Butler et al., 2008). The carbon dioxide emissions are very localized, coinciding with major industrial cities and are mostly due to energy use and industrial processes. The distribution of methane emissions also has

significant contributions from cities, but agriculture is a large source as well. Nitrous oxide emissions have a higher agricultural component still and hence small correlation with city locations. According to the EDGAR 4.0 emission database (EDGAR, 2009), the highest CO_2 emission contributing cities are New York (700 Tg/year), Shanghai (280 Tg/year) and Tokyo (270 Tg/year). In Europe, they are the Ruhr urban area (200 Tg/year), London (90 Tg/year) and Paris (50 Tg/year) based on SDR (2019). CO_2 emission by Prague in Czech Republic is around 2.2 Tg/year for year 2019 CENIA (2019).

The second family of species emitted by cities is comprised of short lived gases and aerosols. They remain airborne for timescales ranging from seconds to months, because they are often highly reactant or undergo efficient removal processes resulting in deposition on earth surface. The most important are the oxides of nitrogen (NOx), that are produced in North American and European cities mainly due to fossil fuel combustion in road transportation and energy production. Carbon monoxide (CO) is product of incomplete combustion and is dominantly emitted in African and Asian cities reflecting the older-than-average technologies used (Streets and Waldhoff, 2000). Non-methanic volatile organic compounds (NMVOCs) are product of road transport and solvent use in North American and European cities; however in Africa and Asia, they originate mainly from domestic combustion Denier van der Gon et al. (2010). SO_2 emissions are released mainly due to energy production and industry and they are low in European cities and higher in North America, Africa and Asia.

Cities emit also aerosols. The most important are carbon containing aerosols: black and organic carbon (BC and OC). They are formed during combustion processes: in developed countries mainly within road transportation, in less extent from industry and domestic/commercial combustion (Denier van der Gon et al., 2010). Here we need to emphasis, that gaseous precursors emitted by cities result in large burden of secondary inorganic and organic aerosols that will be discussed later.

In the Czech capital, Prague, considering emissions of aerosols (or particulate matter, PM), NOx, CO, NMVOCs, SO_2 and ammonia (NH_3), CO emissions dominate as well with 45 % contribution to the total country emissions (11045 Mg/y in absolute numbers). Then NMVOC, NOx, SO_2 and NH_3 emissions follow with 30 % (13158 Mg/y), 3.7 % (6314 Mg/y), 4.3 % (5877 Mg/y) and 0.6 % (398 Mg/y) contribution, respectively (CHMI, 2014). Mobile sources are the main contributors to the overall emissions of CO, NOx and NH_3 with 89 %, 64 %, 95 %, respectively. PM emissions make 5 % of all Prague emissions with 67 % mobile source contribution. The main source of NH_3 is agriculture (95 % in Europe) and waste treatment (1.9 %) that do not take place in cities in considerable amounts according to the European Environmental Agency.

1.2.1 Impact of urban emissions on atmospheric chemistry

Short lived gases and particulate matter released by cities can perturb the background atmosphere chemical composition significantly not only over the cities themselves but, according to the geographical and climatic conditions, at much larger distances (Lawrence et al., 2007) and even on global scale (Timothy et al., 2009).

Emissions of predominantly nitrogen oxides and VOCs are affecting photochemistry. A solely NOx environment leads to photostacionary levels of troposheric ozone. First the nitrogen dioxide (NO_2) is photo-dissociated into nitrogen monoxide (NO) and atomic oxygen. The later reacts with molecular oxygen resulting in ozone (O_3). The cycle is closed by the reaction of O_3 with NO forming NO_2. When VOCs are present, they are transformed to peroxyradicals ($RO_2^{\cdot}$) that enter the cycle above by reacting with NO yielding NO_2. This prevent the destruction of ozone leading to its accumula-

tion (Wayne, 2000). When the concentrations of NOx are much higher than of VOCs (NOx-saturated case), the ozone formation is controlled by the changes of VOCs: ozone increases with increasing VOCs while if NOx increases, ozone decreases by titration. This regime is called VOC-controlled. On the other hand when VOCs/NOx ratio is high, ozone production depends on the change of nitrogen oxides: with increasing NOx concentration ozone increases as well and a NOx-controlled regime occurs (Sillman, 1999).

Cities emit large amounts of both NOx and VOCs. These emissions can lead to both regimes depending on the location with respect to the city, the meteorological conditions and the actual ratio of NOx and VOCs emissions. This ratio is usually high in North-American agglomerations, many eastern Asian cities and in European agglomerations like Athens, Paris, Milan or Berlin (Beekmann and Vautard, 2010; Huszar et al., 2016a). Ozone is usually titrated over these cities. However, according to actual meteorological conditions, pollution from cities can be transported at large distances in the form of odd oxygen (O_x=$O_3 + NO_2$) where the plume from the city mixes with additional VOCs sources and can become NOx sensitive leading to ozone production (Beekmann and Derognat, 2003).

Although they not represent a major source within urban environment, VOCs of biogenic origin (BVOC; Simpson et al., 1995) can significantly affect the regimes at certain cities. While, as already said, in the city's center ozone is titrated due to intensive NOx emissions consuming the hydroxyl radical (OH), on its suburban quarters the NOx can mix with the BVOC resulting in high production of ozone (Holoubek et al., 2005). This occurs mainly in cities surrounded by large forested areas (e.g. Kleinman et al., 2005).

Carbon monoxide and methane can play role in ozone production over/near cities as well, although their effect is believed to be rather small. CO however remains important due to its harmful effect on human health (Bascom et al., 1996).

Emission of gaseous pollutants from cities can further perturb the aerosol burden. This happens not only locally, but it can influence regional scales (de Sá et al., 2018). Sulfur dioxide, nitrogen oxide and ammonia emissions lead, in presence of water vapor, to formation of secondary inorganic aerosols: ammonium-sulfate-nitrate particles (Martin et al., 2004). The primary precursor for sulfate aerosol formation is sulfur dioxide. Barth and Church (1999) investigated the sulfate formation due to SO_2 originating from Mexico City and cities from southeastern China, still the largest SO_2 emitter regions nowadays, and they found significant perturbation of the global sulfate aerosol burden due to these two regions and cities located therein. Further, NOx emission does not affect only photochemistry (and the consequent ozone formation/destruction) but also the formation of nitrate aerosol. If the meteorological conditions are favorable, nitrate oxide emissions from cities can enhance background nitrate aerosol levels significantly (Lin et al., 2010).

Emissions of ammonia from cities are an efficient contributor to formation of sulfate and nitrate aerosol (by forming ammonium-sulfates and ammonium-nitrates) and its importance in connection with cities emissions are studied recently by many studies (Behera et al., 2010, and references therein). Generally, the thermodynamic system of ammonium-sulfate-nitrate-water solution is rather complicated and its equilibrium state is highly dependent on the initial ratio of the $SO_2-NOx-NH_3$ emissions and the governing meteorological conditions (Martin et al., 2004), thus the contribution of different cities to these particles can be very variable.

Finally, organic gaseous material (volatile, intermediate- and semi-volatile VOC) released from cities can contribute to formation of secondary organic aerosols and significantly enhance the total aerosol burden in urban environment as showed by Paredes-

Miranda et al. (2009) or Hodzic et al. (2010).

Air pollution caused by cities has been subject to large measurement campaigns in the last decade (e.g.; Moline et al., 2007; Molina et al., 2010). Measurement data help to characterize the chemical nature (their impact on atmospheric chemistry) and the quantity of these pollutants and represent an essential constrain for air-quality models they are often affected by systematic biases.

The complex nature of chemical interaction of concentrated emissions sources with the background pollution levels requires to adopt numerical modeling techniques when analyzing the urban emission contribution to air-pollution. As one of the first such studies, in Section 3 we present our results using a regional chemistry transport model to investigate the impact of emission from cities on the regional tropospheric chemistry. In Section 9, we add new results showing that for such calculations, one must correctly include the meteorological conditions over urban areas, therefor the urban canopy meteorological forcing (see further) has to be properly represented in numerical models of atmosphere.

1.2.2 The urban canopy meteorological forcing

Mechanical, radiative, thermal and hydraulic properties make urban surfaces in cities clearly distinguished from natural surfaces.They therefore represent additional sinks and sources of momentum, heat and moisture affecting the mechanical, thermodynamical, and hydrological properties of local atmosphere and have specific impact on the meteorological conditions (Oke, 1982, 1987; Eliasson and Holmer, 1990; Haeger-Eugensson and Holmer, 1999).

One of the most widely studied aspects of the meteorological impact of urban surfaces is the Urban Heat Island (UHI) phenomenon which represents an excess warmth of urbanized areas with respect to their non-urbanized (rural) vicinity. In general, UHI forms due to significant perturbation of fluxes of energy, moisture and momentum within this environment, which is characterized by canyon-like geometry and specific thermal parameters of the artificial surfaces (Oke, 1982). Due to their decreased albedo, urban surfaces store more heat compared to rural areas and after sunset this heat is released with a reduced efficiency because of the decreased sky-view factor (Grimmond and Oke, 1995) making UHI typical for nighttime. The decreased sky-view is the result of the canyon geometry and causes trapping of both shortwave and longwave radiation. UHI is detectable during daytime as well, however, with a limited intensity. The UHI is further enhanced by several other factors. Increased anthropogenic heat emission within urban environment increases urban temperatures (Block et al., 2004). Further, urban areas covered by impervious surfaces exhibit higher runoff than their rural counterparts, which leaves them with less surface water available for evaporation. Lower evaporation than decreases the latent heat consumption leading to temperature increase (Grimmond and Oke, 1991; Taha, 1997).

Some studies performing measurement in and around cities revealed another phenomenon: the so called Urban Cool Island (UCI) effect – during the morning hours the enhanced shadowing within urban surfaces delays the heating and sometimes causes lower temperatures than over the rural surfaces (Basara et al., 2008; Gaffin et al., 2008).

Urban surfaces have further impacts on other meteorological parameters as well: Richards and Oke (2002) and Richards (2004) studied the changes of surface humidity, while Grimmond and Oke (2002), Roth (2000) and Kastner-Klein et al. (2001) focused on the impact on roughness and turbulence. Many studies dealt with the structure of the urban boundary layer including the impact on the height of the planetary boundary layer (ZPBL) (Piringer, 2001; Martilli, 2002; Angevine et al., 2003; Nair et al., 2004)

and wind speed (Hou et al., 2013). Urbanization-triggered changes in precipitation and hydrological processes got as well into the attention of research (e.g. Shepherd et al., 2002; Rozoff et al., 2003). There is evidence that the urban environment with its higher air pollution is responsible for enhanced lightning (Coquillat et al., 2013). Schaldach and Alcamo (2007) showed significant influence on the carbon balance as well and, finally, the urban-meteorology interaction may significantly influence air-quality (Ryu et al., 2013a,b; Huszar et al., 2018). However, most of the influences listed here have to be viewed in a common UHI-related framework as they are all physically connected within this phenomenon, bringing higher street level temperatures and having a direct impact on the human health (Reid et al., 2009) and, in general, on the comfort of living.

In summary, meteorological conditions are strongly modified due to the rural-to-urban transitions and therefor, the urban land surfaces can be treated as a forcing and we will call this forcing as urban canopy meteorological forcing (UCMF), as we introduced in Huszar et al. (2020).

The complex nature of urban canopy influence on the meteorological conditions which includes many features makes modelling of this phenomenon very important and understanding them makes the use of modelling tools inevitable. Here, of course, higher resolution models that directly simulate the evolution of the state of the air urban street canyons bring the most detailed picture (like CFD - Computational fluid dynamics; Mirzaei and Haghighat, 2010). These high resolution approaches are however are computationally very demanding and inappropriate for use in long-term climate related regional scale studies. Another possibility is to use meso-scale models which have horizontal resolution approximately ranged from one to several-hundreds of kilometers. These are less demanding on computer resources and are thus useful for climate scale studies of urban canopy interaction with air. Current mesoscale models (numerical weather prediction as well as regional climate models) still fail to capture properly the impact of local urban features on the mesoscale meteorology and climate, despite of their increasing resolution. Therefore the inclusion of urban-canopy-models (UCM), which are specially designed to parameterize the processes specific to the urban environment that are not resolvable at the modelś scale, is necessary (Baklanov et al., 2008; Lee and Park, 2008; Oleson et al., 2008; Chen et al., 2011). UCMs enables to account for the small and microscale urban effects over much larger scales (from kilometers to hundred of kilometers) and thus to investigate the impact of cities on regional and global scale weather and climate.

In Section 2, we present our results, as one of the first studies concerning the long term impact of urban canopy forcing on meteorological and climate conditions for the region of wider central Europe. Using a more advanced UCM, more pronounced results on the meteorological modifications are presented also in Section 5 and 6, while in Section 7 the attention is focused on the modification of the vertical eddy diffusion.

1.3 Indirect impact of urban areas on atmosphere

This section brings to reader some concepts about the indirect impact of cities on the atmosphere which includes the interaction of urban emissions with solar and terrestrial radiation and the impact of urban canopy meteorological forcing on air chemistry. First, the general pathways of interactions between meteorological conditions and climate at one side, and atmospheric chemistry on the other side, are investigated.

1.3.1 Interactions between meteorology/climate and atmospheric chemistry

The atmosphere is a complex system with several subcomponents that are in continuous interaction. The horizontal and vertical motion of air is governed by atmospheric dynamics, formation of cloud an precipitation and the hydrological cycle in general is described by cloud-/rain-microphysics (microphysics in brief) and, finally, atmospheric radiation subjects the penetration of solar and terrestrial radiation trough different layers of air. These are further extended by chemical processes acting in air on both gaseous material and particles (aerosols) emitted to or formed in the atmosphere. The first three subsystems can be considered to determine the meteorological conditions and, if averaged over sufficiently long period, the climate. The last two subsystems are, on the other hand, traditionally treated within atmospheric chemistry. To complete the picture, one has to consider also the "boundary systems" that serves as lower, upper or side boundary conditions to the listed components, like ocean dynamics, cryosphere (e.g. the physics of sea-ice and land-ice) and biosphere (containing both natural and anthropogenic influences).

Between meteorological/climate conditions and atmospheric chemistry, many pathways of mutual interaction act. Fig. 1.1 highlights the most important ones. These contains links between subsystems within meteorology: dynamics, radiation and microphysics, as well as links between subsystems within atmospheric chemistry: gaseous species and aerosols. Here, we will however focus on the interactions between three "meteorological" components and the two "chemical" components.

Starting with the influence of meteorological conditions on chemistry, let's focus first on the impact of atmospheric dynamics. Horizontal and vertical motions of air directly impact the transport of chemical species. Convection leads to adiabatic cooling of air leading to lower temperature that impacts the temperature dependent reaction rates or nucleation of gases into aerosols. In climatic scales, changes in wind direction or speed can directly influence air-quality by changing pollutant transport (Huszar et al., 2011; Langer et al., 2012). Further, changes of air temperature in changing climate can largely influence the formation of secondary pollutants like ozone as well as deposition velocities (Katragkou et al., 2011; Kolozsi-Komjáthy et al., 2011). Temperature changes can also alter photolysis rates which, besides wave-length, depend also on ambient temperatures. Another pathways include impacts of turbulence and their changes on the mixing and dispersion of pollutants as well as on the enhancement of coagulation of aerosol particles.

Secondly, regarding the impact of atmospheric radiation, the most important pathway is the influence via the modifications of photolysis rates. Their changes can be a result of many reasons: changes in cloudiness, ozone column, aerosol optical depth etc, and any change in their value directly induce changes in photochemistry impacting pollutant concentrations (Xing et al., 2017). In case of aerosols, there is an evidence that photolysis can act in aqueous phases as well changing the chemical structure of aerosols (Epstain et al., 2013). Further, changed photolysis rates in future impacts the emissions of organic species that are precursors for organic aerosol formation (Jiang et al., 2010).

Thirdly, the presence of clouds and precipitation impacts the aqueous chemistry in air, i.e. chemical processes involving water droplets and ice. Changes in cloud droplet characteristics (number concentration, size spectra etc.) influences the water uptake of many gases with moderate to strong solubility and decreasing their gas phases concentrations. Enhanced/reduced liquid water content further increases/decreases the wet deposition rates in future under changing climate (Simpson et al., 2014).

In the opposite direction the atmospheric chemistry, or more specially the spatial

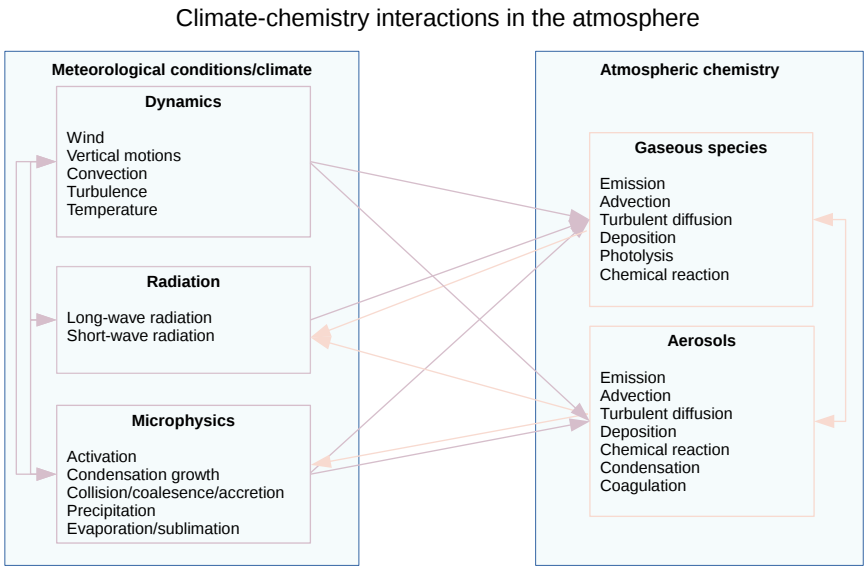

Figure 1.1: Interaction pathways between climate and atmospheric chemistry.

and temporal distribution of the chemical composition of the atmosphere affect many meteorological processes. There are two primary influences: 1) the direct impact of atmospheric gases and aerosols with solar and terrestrial radiation, and 2) the impact of aerosols on formation and evolution of clouds and rain.

The most important aspect of the direct impact on radiation is the role of radiatively active gases. These absorb the solar radiation at different wavelengths (molecular oxygen, water vapor, ozone or carbon dioxide). Changes in their concentration is immediately reflected in amount of radiation absorbed which in turn influences the thermal balance of the relevant atmospheric layers. Here, the most widely studied link is between the changes of stratoshperic ozone and stratospheric temperatures. Consequently, changes in stratospheric temperatures, induced by changes in ozone, can alter the whole dynamics of the stratospheric circulation (Butchart et al., 2006). There are also atmospheric gaseous species absorb long wave infrared radiation penetrating the atmosphere either from the the earth or originating from the air itself above (including heterogenic bodies like clouds). These gases, called green-house-gases, act as a shield preventing infrared radiation from earth to escape to space by re-emitting it towards the surfaces. The most important GHGs are carbon dioxide (CO_2), ozone (O_3, methane CH_4 or nitrous oxide N_2O). Changes in their concentrations alter the radiative and thus thermal balance of the air potentially leading temperature changes. Indeed, the global warming observed is largely attributed to the increased burden of GHGs (IPCC, 2013). Among green-house-gases, ozone represents a relatively complicated role as it has a short lifetime a thus high spatio-temporal variability. For example, Park et al. (2001) showed that under favorable conditions, tropospheric ozone can accumulate near the sources to such levels that it will trigger detectable local and regional warming.

Besides radiatively active gases, aerosols interact with radiation too: they scatter shortwave direct and diffuse solar radiation - this include reflection too (Martin et al.,

2004). Further, if aerosol have a low reflective surface, it has the capability of absorbing shortwave radiation. Black carbon is a typical representative of such aerosol and many studies showed its role in modulating the radiative and thus thermal balance in the troposphere (Huszar et al., 2011; Dumka et al., 2013). Consequently, any changes (e.g. in future given by changed emissions) in aerosol concentration hav the potential to perturb the radiative hence thermal balance of air leading to temperature changes (Levy et al., 2013).

As already mentioned, incoming solar radiation can be scattered by aerosols resulting in a "direct effect" associated with a cooling at the surface. Incoming solar radiation also can be absorbed by aerosols consisting of black carbon and mineral dust, heating the local atmosphere and possibly reducing the incidence of cloud formation through the "semi-direct effect" (Johnson et al., 2004) – an often investigated phenomenon regarding marine stratus cloudiness. With the ability to promote water drop nucleation or freezing, aerosol can act also as cloud condensation nuclei (CCN) and/or ice nuclei (IN). Such aerosols may affect cloud microphysics by increasing the number of droplets and cause a decrease in droplet size for fixed liquid water content. This thus influences the overall cloud radiative properties through interactions referred to as the "first indirect effect" (Krüger et al., 2002). The first indirect effect pushes the drop spectra towards smaller sizes which in turn reduces the collision efficiency and coalescence of droplets leading to precipitation suppression. Reduced precipitation than can prolong the cloud lifetime and thickness, the process knowns as the "second indirect effect" (Lohmann and Feichter, 2005). Changed cloud properties (including its lifetime) than directly impacts the radiative balance.

The above summarized interactions between climate and atmospheric chemistry become interesting above areas where one of the subsystems (either climate or atmospheric chemistry) is significantly perturbed with effects in the other subsystem. Cities represent such areas because they are 1) characterized by intense concentrated emissions leading to great perturbation of background air-pollution and 2) are covered by artificial materials with specific geometry substantially modifying the meteorological/climate conditions. It is thus clear that climate-chemistry interactions in the Earth atmosphere should receive special attention over urbanized areas.

1.3.2 Impact of urban emissions on meteorology and climate via radiation

As already mentioned, cities emit large amount of both short and long-lived materials into air substantially altering the chemical composition not only on local, but also on regional and even global scale (e.g. Lawrence et al., 2007; Folberth et al., 2010; Markakis et al., 2015). Due to the fact that many of these gases/aerosols interact with solar or/and longwave radiation, these chemical perturbations must lead to perturbation of radiative hence thermal balance.

While there is a large number of studies that focused on the impact of short lived pollutants from cities, both measurement (Freney et al., 2014; Molina et al., 2010) and modelling based (Li et al., 2011; Finardi et al., 2014; Skyllakou et al., 2014; Markaris et al., 2016), much less work has been done regarding the climate impact of urban emissions and consequent chemical perturbations. Recently, two reviews were published addressing the urban emission impact on climate (among other impacts): Folberth et al. (2015) and Baklanov et al. (2016). Focusing on so called megacities (cities and urban agglomerations with inhabitants larger than 10 million), both agree, that there is high confidence on the climate impact of CO_2 emitted and that much of the radiative impact of urban emission is attributable to this gas. Folberth et al. (2012) estimates the global radiative forcing (RF) to 120.0, 28.4 and 3.3 $\mathrm{mW\,m^{-2}}$, respectively, from

the long-lived components CO_2, methane (CH_4) and nitrous oxide (N_2O) emitted by megacities. For the impact of short-lived species, namely ozone and aerosol, they give a global mean RF 5.7±0.02 mW m^{-2} due to the increase in tropospheric ozone, and -6.1±0.21 mW m^{-2} due to urban induced aerosol increase. Comparing these last two numbers, it is clear that on global average, ozone and aerosol climate effects nearly cancel each other.

On local and regional scale, however, the impact of short lived pollutants can be much more important as outlined by (Baklanov et al., 2016). As a result of emissions of oxides of nitrogen and hydrocarbons, ozone is produced and under favorable weather conditions it can accumulate over and around cities. Consequently, being green-house-gas, it can trigger positive radiative forcing that results in warming (Park et al., 2001). Many studies showed significant effect of aerosol originating from cities or highly populated agglomerations on radiation and consequently meteorology and climate. Black carbon and its radiative feedbacks from an Indian megacity was investigated by Tripathi et al. (2005) – in their study black carbon underwent short wave atmospheric absorption which translated to a lower atmospheric heating of $-1.8K/day$. Giorgi et al. (2002) attributed observed cooling in a highly populated area in China to sulfate and carbonaceous aerosol that originated from the same area. Ramanachan and Kedia (2010) looked at black carbon pollution over Ahmedabad, India and indicated its important role in the radiative budged possibly affecting even the monsoon rainfall. At last, the radiative impact of material emitted from cities is found to depend also on the age of the associated plume: Roldin et al. (2011) calculated the radiative impact of aging urban plumes from Malmö, Sweden and found a RF from -0.3 to -3.3 mW m^{-2} depending on the distance from city and the specific cloud properties.

We contributed to the mentioned papers by evaluating the long term (10yr long) impact of urban emissions and consequent chemical perturbations on the radiative balance and climate over central Europe. Our results are presented in Section 4.

1.3.3 Impact of urban canopy meteorological forcing on atmospheric chemistry

As detailed earlier, cities strongly modify the mechanical and thermodynamical properties of air above, as they are substantially different than their natural counterparts by the properties of the materials they are covered the geometry of urban features (like buildings, streets etc.). As meteorology and ,thus, climate is strongly influences air quality and atmospheric chemistry in general, it is straightforward to expect that due to urban canopy induced meteorological changes (i.e. due to UCMF) chemistry will be modified as well.

The most profound modification cities bring is the formation of the urban heat island, which brings higher temperatures in cities than over their vicinity. Elevated urban temperatures increase reaction rates enhancing chemical reaction speed. 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/to urban areas depending on the time of the day (Ryu et al., 2013a; Hidalgo et al., 2010). However, there is an opposite effect caused by the drag associated by the urban canopy: the drag induced wind stilling can slow down pollution dispersion into larger scales, for both primary pollutants and secondary formed ones. The high variability of urban surfaces further increase turbulence and causes stronger eddy-diffusion which supports vertical transport of pollutants away from the urban canopy layer. This often leads 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). There are also notable effect of the urban induced rain pattern change,

which is especially evident in connection with convection (Rozoff et al., 2003). Changed spatial patters of precipitation consequently perturbs the pollutant concentrations by spatial modifications in wet deposition.

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. Enhanced vertical turbulence, for example, 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. Another example is related to elevated temperatures, which 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 they can contribute to their more effective destruction, like in case of NOx-ozone interaction (formation vs. titration).

Secondly, the urban induced meteorological changes are not uniform in time and have a very specific daily cycle (Gaffin et al., 2008). 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 became clear that the use of integrated modeling tools for both meteorology and air-chemistry is a necessity in analyzing the impact of urban canopy induced meteorological changes on air-chemistry and many studies appeared that evaluate the magnitude of this impact over regional scales, focusing usually on the short term effects rather than looking at the long term impact (e.g. Wang et al., 2009; Struzewska and Kaminski, 2012; Ryu et al., 2013a, and others).

Our first study in this regard, presented in Section 5, focuses rather on the long term impact of the urban canopy forcing on air-quality and it arrives to conclusions about how important are the different processes characteristic to urban environment in modulating urban air pollution while gas-phase species are analyzed. Section 6 present a similar study but focusing on aerosol modifications, while Section 7 details the fundamental role of atmospheric turbulence in the UCMF modulated air-quality changes and here, the term "Urban Canopy Meteorological Forcing" is explicitly defined for the first time. The question, whether the impact of UCMF is more pronounced for extreme air pollution is answered in Section 8 and, finally, Section 9 shows that the regional footprint of urban emissions is greatly modulated by the UCMF which thus cannot be neglected in urban oriented air-quality studies.

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List of papers

The presented thesis entitled *Impact of urbanization on climate and atmospheric chemistry* is compiled from the following peer-reviewed publications:

Huszár, P., Halenka, T., Belda, M., Zak, M., Sindelarova, K., and Miksovsky, J.: Regional climate model assessment of the urban land-surface forcing over central Europe, Atmos. Chem. Phys., 14, 12393-12413, doi:10.5194/acp-14-12393-2014, 2014.

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Huszár, P., Belda, M., Karlický, J., Pišoft, P., and Halenka, T.: The regional impact of urban emissions on climate over central Europe: present and future emission perspectives, Atmos. Chem. Phys., 16, 12993-13013, doi:10.5194/acp-16-12993-2016, 2016b.

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2. Regional climate model assessment of the urban land-surface forcing over central Europe

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3. On the long term impact of emissions from central European cities on regional air-quality

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4. The regional impact of urban emissions on climate over central Europe: present and future emission perspective

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5. The impact of urban canopy meteorological forcing on summer photochemistry

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6. Impact of urban canopy meteorological forcing on aerosol concentrations

This chapter is based on the author's study in Atmospheric Chemistry and Physics: *Huszar, P., Belda, M., Karlický, J., Bardachova, T., Halenka, T., and Pisoft, P.: Impact of urban canopy meteorological forcing on aerosol concentrations, Atmos. Chem. Phys., 18, 14059-14078, https://doi.org/10.5194/acp-18-14059-2018, 2018b.*

7. Urban canopy meteorological forcing and its impact on ozone and PM2.5: role of vertical turbulent transport

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8. The impact of urban land-surface on extreme air pollution over central Europe

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9. The regional impact of urban emissions on air quality in Europe: the role of the urban canopy effects

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10. Summary and Outlooks

The impact of cities and, in general, urban surfaces on climate and atmospheric chemistry was analyzed over Europe using regional scale coupled climate-chemistry models. Direct and indirect pathways of urban influence on atmospheric environment were identified and studied. More specifically, the direct impact of urban surfaces on meteorological conditions and climate, the direct impact of urban emission on tropospheric chemistry, the indirect impact of urban emission-induced chemistry perturbations on atmospheric radiation and climate and, lastly, the impact of urban canopy induced meteorological forcing on tropospheric chemistry were quantified, while special attention was dedicated to the role of vertical turbulent transport and to extreme air pollution too.

The impact of urban surfaces on climate is characterized by a clear increase of temperatures by up to 1-2 K over cities as summer average. From a diurnal cycle view, the impact is most pronounced during evening hours, reaching 3 K. There is a statistically significant impact over rural areas, indicating some minor remote influence of cities. The impact on wind speed is characterized by decreases up to 1 ms^{-1}, peaking over cities and during noontime. However, the impact differs between the applied urban canopy parameterizations used within the driving model. The more advanced CLM4.5 model used in Huszar et al. (2018a,b) (and in our later studies) tends to predict, and this is also true for temperature, a more stressed urban impact on wind than the SLUCM model that we implemented in Huszar et al. (2014).

The urban generated turbulence was subject of the analysis of Huszar et al. (2018a) and Huszar et al. (2020a), where it was found that the additional drag introduced by urban surfaces and the temperature-induced reduction of stability increased the vertical turbulent diffusion by 60–70 m^2s^{-1} over cities. This increase is closely connected to the modifications of the boundary layer height, which is increased by more than 100 meters due to urban surfaces, especially during summer.

Urban surfaces represent strong sinks in the hydrological cycle with high value of run-off due to sewer systems and drainage and low value of evaporation. This causes negative impact of urban surfaces on water vapor content. Indeed, we simulated decreases of water vapor mixing ratio over cities by up to 1 $g.kg^{-1}$. Low evaporation is also responsible for suppressed precipitation over cities by up to few mm per day.

A strong impact on regional scale air quality due to urban emission was simulated too (Huszar et al., 2016a). Significant ozone titration occurred over cities while over rural areas remote from cities, a net ozone production was modeled, mainly in terms of number of exceedances and accumulated exceedances over the threshold of 40 ppbv. It was further found that urban NO_x, SO_2 and $PM_{2.5}$ emissions also significantly contributed to concentrations in the cities themselves (up to 50–70 % for NO_x and SO_2, and up to 60 % for $PM_{2.5}$), but the contribution is large over rural areas as well (10–20 %). Although air pollution over cities is largely determined by the local urban sources, considerable (often a few tens of %) fraction of the concentration is attributed to other sources from rural areas and minor cities. For the case of Prague, it was further shown that the inter-urban interference between large cities does not play an important role which means that the impact on a chosen city of emissions from all other large cities is very small. At last, it was shown that to achieve significant ozone reduction over cities in central Europe, the emission control strategies have to focus on the reduction of NMVOC, as reducing NO_x (due to suppressed titration) leads often to increased O_3. The influence over rural areas is however always in favor of improved air-quality, i.e. both NO_x and/or NMVOC reduction ends up in decreased ozone pollution, mainly

in terms of exceedances.

As expected, the simulated changes in species concentrations may result in the perturbation of radiative balance, if these species interact with radiation. Indeed, this is the case for ozone and for aerosol and statistically significant impact on climate was modeled (Huszar et al., 2016b). It is characterized by a statistically significant cooling up to -0.02 K and -0.04 K in winter (DJF) and summer (JJA) season, mainly over cities. We found that the main contributor to the cooling is the aerosols direct and indirect effects, while the ozone titration, calculated especially for DJF, plays rather a minor role. In accordance with the vertical extent of the urban emission induced aerosol perturbation, cooling dominates the first few model layers up to about 150 m in DJF and 1000 m in JJA. We also found a clear diurnal cycle of the radiative impacts with maximum cooling just after noon (JJA) or later in afternoon (DJF). Furthermore, statistically significant decreases of surface radiation are modeled, in accordance with the temperature decrease. The impact on the boundary later height is small but statistically significant and reaches 1 m and 6 m decreases in DJF and JJA, respectively. We did not find any statistically significant impact on precipitation and wind speed. This indicates, at least for the region in focus, that regional climate studies concerning the impact of urban environment on the atmosphere should more focus on the impact of urban canopy on the meteorology and climate where the magnitude of the influence was 2 orders higher. However, it must be stressed, that while the climate impact of urban emissions is showing to be rather small, the impact on air-quality remains significant and has to be considered in future.

As air-quality and atmospheric chemistry in general is closely connected to the meteorological conditions, it is clear that the simulated urban canopy induced changes in meteorology/climate have implications in the concentration of pollutants and will act as a certain forcing. We introduced it as the urban canopy meteorological forcing (UCMF) in Huszar et al. (2020a) and, indeed, we simulated statistically significant changes in NOx, HNO_3 and ozone concentrations over cities due to UCMF, more specifically due to temperature-, wind- and turbulence effects detailed above (Huszar et al., 2018a). 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. In Huszar et al. (2018b) we showed that due to the UCMF, aerosol concentrations are significantly decreased (by -1.5 to -2 μgm^{-3}), again mainly due to enhanced vertical turbulent diffusion over urban areas. As these studies looked at impacts on average values, we were interested in Huszar et al. (2020b) whether UCMF impacts extreme air pollution in a more pronounced way. Indeed, we found that this is true for NO_2 and PM2.5. A special attention was dedicated to the impact of UCMF on vertical eddy (turbulent) diffusion by calculating the eddy diffusivities using a range of different diagnostic methods. Huszar et al. (2020a) found that turbulence remains the dominant components of the the total UCMF impact on chemistry over urban areas regardless of the choice of the method. Finally, knowing the UCMF modulation of air-quality over cities, we showed that this has strong implications on the impact of urban emissions on local and regional scales: more specifically, this impact is smaller by about 30-50% if UCMF is considered (Huszar et al., 2021), while the dominant reasons is the above mentioned enhanced vertical eddy diffusion.

The above summarized effects have been quantified at relatively coarse, 10 km x 10 km (eventually 9 km x 9 km) horizontal model resolution, while we used a nested 1 km resolution domain only for Prague (in Huszar et al. (2020a,b, 2021)). The coarse resolution mentioned is certainly sufficient for regional scale studies (Tie et al., 2010; Varghese et al., 2011), however, the fine scale character of urban-climate-chemistry in-

teractions cannot be resolved with this grid step and it is expected that many subgrid effects are disregarded in this fashion. The characteristic length scale of weather phenomenon related to urban effects (urban heat island, impact on turbulence, wind etc.), the transport of urban plume shortly after release and its interaction with urban air via radiation and cloud microphysics cannot be resolved properly with coarse resolution models having several km grid steps. This motivates recent research to apply higher resolution for regional studies going below 1 km x 1 km (Chen et al., 2014, 2016; Göndöcs, 2016) and this was the motivation for us too to apply, at least for one selected city, higher resolution. At these scales, the hydrostatic approximation to atmospheric dynamics in modelling is no longer correct and vertical accelerations have to be accounted for in order to resolve non-hydrostatic dynamics. Vertical accelerations are particularly important in deep convection which penetrates the significant part of the model's vertical extent. With non-hydrostatic approach and the mentioned high resolutions, models are permitted to explicitly simulate convection and they are called, accordingly, convection permitting models (CPM). The main motivation for CPM is "when deep convection is a dominant process (e.g., tropics, subtropics, and mid latitude summer) and in regions with strong spatial heterogeneities (e.g., mountainous regions, coastlines, and urban areas)" (Prein et al., 2015). Further, the model resolutions needed for convection permitting approach are high enough to resolve urban heterogeneities with grid-boxes completely covered with urban-type surfaces and therefor there is a strong added value of applying CPMs simulations to urban regions. Especially, the magnitude of the urban heat island is found to be larger at the CPM scale compared to coarse non-CPM approaches (Wouters et al., 2013). Indeed, even in our results it can be clearly identified, that the impacts over Prague at high resolution are much more pronounced and more detail is provided than using a coarse resolution for horizontal grid-step. This all determines the future research aiming to qualify and quantify the cities' interaction with the atmospheric environment: definitely, higher resolution convection permitting models/configurations have to be applied for larger areas encompassing another large urban areas. In summary, higher resolution brings better representation of the surface heterogeneities by which urbanized areas are characterized. Secondly, higher resolution enables (as already mentioned) the explicit treatment of convection which is believed to be greatly perturbed by urban surfaces, and thirdly, high resolution helps to resolve the detailed nature of emissions, urban air-chemistry and transport of pollutants.

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