Aerosols, Clusters, Greenhouse Gases, Trace Gases and Boundary-Layer Dynamics : on Feedbacks and Interactions

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2023-03

RESEARCH ARTICLE

Aerosols, Clusters, Greenhouse Gases, Trace Gases and Boundary-Layer Dynamics: on Feedbacks and Interactions

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Abstract

Turbulence is the key process transporting material and energy in the atmosphere. Furthermore, turbulence causes concentration fluctuations, influencing different atmospheric processes such as deposition, chemical reactions, formation of low-volatile vapours, formation of new aerosol particles and their growth in the atmosphere, and the effect of aerosol particles on boundary-layer meteorology. In order to analyse the connections, interactions and feedbacks relating those different processes require a deep understanding of atmospheric turbulence mechanisms, atmospheric chemistry and aerosol dynamics. All these processes will further influence air pollution and climate. The better we understand these processes and their interactions and associated feedback, the more effectively we can mitigate air pollution as well as mitigate climate forcers and adapt to climate change. We present several aspects on the importance of turbulence including how turbulence is crucial for atmospheric phenomena and feedbacks in different environments. Furthermore, we discuss how boundary-layer dynamics links to aerosols and air pollution. Here, we present also a roadmap from deep understanding to practical solutions.

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1 Introduction/Background

The planetary boundary layer (PBL) is a key compartment in our globe: we are living inside
it, most of activities as well as pollutants are located there, and interactions between the
atmosphere and the Earth surface occur within the PBL. Therefore, it is crucial to understand
boundary-layer dynamics. The boundary-layer dynamics and vertical structure have been
investigated for several decades using various methods (e.g. Bornstein 1968; Willis and
Deardorff 1974; Seibert et al. 2000; Emeis et al. 2004; Calhoun et al. 2006; Grimmond 2006;
Baklanov et al. 2011; Barlow et al. 2011). Zilitinkevich et al. (2013, 2021) have further
developed rigorous ways to investigate boundary-layer dynamics, particularly the stability
of the PBL. Turbulence is the key phenomena in the PBL (e.g. Kaimal et al. 1972; Roth
2000; Wood et al. 2010). On one hand, turbulence transfers heat and material and decreases
concentration gradients, while on the other hand it causes concentration fluctuations and, in
this way, strengthens several processes, including chemical reactions, new particle formation
(NPF), aerosol growth and activation of cloud condensation nuclei (CCN) to cloud droplets.

Already in 2012, Zilitinkevich et al. (2012) underlined that an unstably stratified PBL
interacts with the free atmosphere mainly through turbulent ventilation at the PBL upper
boundary. This mechanism, still insufficiently understood and poorly modelled, controls the
development of convective clouds, dispersion and deposition of aerosol particles and gases.
Besides unstable PBL, turbulence in a stable PBL has been widely studied (e.g. Smedman
1988; Mahrt 1999, 2014; Galperin et al. 2007). Zilitinkevich et al. recognized the importance
of turbulence in the stably stratified PBL and contributed to its theoretical description (Zil-
itinkevich and Calanca 2000; Zilitinkevich 2002; Zilitinkevich et al. 2007a,b, 2013, 2019;
Zilitinkevich and Esau 2005, 2007). Zilitinkevich and Esau (2009) showed that shallow, stably
stratified PBLs, occurring in winter and being sensitive to weak environmental changes, are
typical in Northern Scandinavia and Siberia. They also noted that winter-time, high-latitude
PBLs need to be studied especially under conditions of environmental and climate changes.

In a bigger picture, PBL is a place for land–atmosphere–ocean processes and interactions
(e.g. Delage and Taylor 1970; Schiermeier 1978; Menut et al. 2000; Arnfield 2003; Lemonsu
et al. 2006; Barlow et al. 2011; Ding et al. 2016). A better understanding of global atmo-
sphere–earth interactions builds on analyses, observations and monitoring of a wide range
of parameters, including the PBL characteristics. The need for a comprehensive inventory
of the PBL dynamics over Eurasia was brought up in the Pan-Eurasian Experiment (PEEX)
Science Plan (2015) which states: "*The traditional view of the atmosphere–earth interac-
tions as fully characterized by the surface fluxes is incompatible with the modern concept of a
multi-scale climate system, where PBLs couple the geospheres, and regulate local features of
the climate. In this context the PBL height and the turbulent fluxes at the PBL upper boundary
play important roles*". In general, the PEEX programme focuses on the Northern Eurasian
arctic–boreal region and on urban environments in China (Kulmala et al. 2015, 2016; Lap-
palainen et al. 2022a; PEEX 2015). The PEEX is especially interested in quantification of
the land–atmosphere climate interactions in arctic–boreal environment (e.g. Kulmala et al.
2020), and in solving complex air quality processes in Chinese mega cities (e.g. Kulmala
et al. 2021; Petäjä et al. 2016).

In this paper, we give an overview on the role of turbulence in the dynamics of the PBL, in the understanding of air quality in extreme polluted environments and in addressing some important land–atmosphere feedbacks, referring to S. Zilitinkevich's fundamental theories and contribution to atmospheric sciences and his studies on PBL. We first summarize (chapter 2) the importance of turbulence and give some examples how fluctuations due to turbulence will affect aerosol nano- and microphysics as well as chemistry. Then we discuss (chapter 3) how boundary-layer dynamics is interacting with air pollution and urban processes. This includes urban heat and pollution islands. In both chapters (2 and 3) we discuss also several feedback loops and interactions. Since there are several open questions to be investigated, we presented in chapter 4 a roadmap to study and solve those questions.

2 On the Importance of Turbulence

Turbulence is generated by both mechanical and thermal processes (e.g. Monin and Zilitinkevich 1969; Zilitinkevich 1970, 2013; Oke 1988; Stull 1988; Sorbjan 1989). The mechanically generated turbulence in the atmospheric boundary layer is routinely formed by the frictional force imposed by the surface roughness elements (e.g. buildings and trees) to the mean flow (wind shear), whereas the thermally generated turbulence is typically resulting from surface heating (buoyancy driven mixing). Mechanically generated turbulent eddies are relatively small and typically scale with the size of roughness elements, whereas the thermally generated eddies scale with the height above the surface and are constrained only by the presence of the ground and depth of the atmospheric PBL. Therefore, the two sources of turbulence result in eddies of different scales and typically both production processes are effective at the same time, so that there is a varying size of eddies in the atmosphere. The relative contribution of either process depends on the roughness and inhomogeneities of the surface, wind speed and the strength of the surface heat fluxes.

Depending on the thermal stratification, one can distinguish between stable, neutral and unstable boundary layers. In a stable boundary layer, turbulence is suppressed yet never completely fades out. Zilitinkevich et al. (2013, 2019) showed that supercritical turbulence makes a selective vertical transport: always pronounced for momentum (maintained by velocity shear), but sharply weakening with an increasing stability for heat and matter. In an unstable boundary layer, it has been commonly believed that the eddies produced by both of the mechanisms break down into smaller ones producing a direct cascade of turbulent kinetic energy and other properties from large to small scales until they are small enough to be dissipated. This process is inherent to the mechanically generated turbulence. Instead, convective plumes can demonstrate a notably different behaviour merging with each other and form self-organized structures, such as cells or rolls often observed in the PBL, e.g. as cloud streets (Zilitinkevich et al. 2021). In his novel theory, Zilitinkevich et al. (2021) reveal that besides a mechanical part of vertical turbulence which moves in all directions and breaks down into smaller eddies making the direct cascade, there is also the convective turbulence composed of vertical buoyant plumes, which merge into larger plumes producing the inverse cascade towards their conversion into large-scale self-organized structures. In this chapter, we discuss the important role that turbulence plays in different environments (on the example of urban boundary layer), phenomena (as an example of new particle formation and growth) and land–atmosphere feedbacks (on the example of COBACC feedback loop).

2.1 Urban Boundary Layer

Due to both mechanically and thermally induced turbulence, the PBL, especially in urban areas, is almost always in a turbulent state (Roth 2000). During nighttime with strong wind and overcast, the atmospheric stability conditions can be neutral (Monin–Obukhov (Monin and Obukhov 1954) stability parameter $\zeta \sim 0$), but stable cases ($\zeta > 0$) are practically almost impossible in urban areas even in nighttime (e.g. Nakamura and Oke 1988; Roth 2000), except at high latitudes. Instead, unstable ($-1 < \zeta < 0$) and very unstable ($\zeta < < -1$) atmospheric conditions are typical in urban areas due to mechanical and thermal turbulence created by the high surface roughness and urban heat island (UHI) phenomena.

In urban climate and pollution studies, it is crucial to take into account turbulence and atmospheric stability conditions in order to understand all interactions and feedbacks between aerosol dynamics and urban climate in different scales. Since influences of smaller, mechanically generated eddies tend to decrease with an increasing distance from the ground (Solanki et al. 2022), and since thermally produced eddies scale in size with the height, the latter are usually more efficient in mixing constituents in the PBL. However, the smaller mechanically generated eddies are important in mixing pollutants close to the surface (Barlow 2014), but also causing the high pollutant concentration gradients due to the turbulence generated by the individual roughness elements (Britter and Hanna 2003). The more we have mechanically induced turbulence, the more there are small-scale spatial variations in pollutant concentrations due to individual roughness elements in urban areas. In addition, mechanically induced turbulence can also improve the ventilation of street canyons and therefore lead to lower near-surface concentrations of pollutants (Zhang et al. 2020). The high variability of, e.g. O_3 affects the oxidation capacity, together with turbulence induced reduction of condensation sink, are influencing new particle formation (NPF) and thereby haze formation in urban areas (Kulmala et al. 2021; Wu et al. 2021). In the presence of strong haze, the intensity of incoming solar radiation is attenuated, which suppresses or prevents NPF. Therefore, we need to understand interactions between the turbulence, haze, radiation balance and NPF to identify the tipping point at which NPF ceases in urban environments.

Turbulence, and especially the presence of large thermally generated eddies, is one of the most important characteristics determining boundary-layer dynamics (Zilitinkevich 2013). However, haze is attenuating the incoming solar radiation, decreasing surface heating and thereby thermally generated turbulence, which cause a decrease in the boundary-layer height (BLH) (Petäjä et al. 2016; Liu et al. 2018). Different aerosol types attenuate the incoming solar radiation with different magnitude, in addition to which PBL dynamics is strongly influenced by the vertical distribution of absorbing aerosols such as black carbon (e.g. Ding et al. 2016). The decreased BLH in turn decreases the volume of air into which pollutants accumulate, which further decreases the BLH. Similarly, the UHI-induced circulation is bringing cleaner air into the city from surrounding rural areas, but with decreased surface heating due to the haze, the UHI intensity and therefore also the UHI circulation will be decreased. Therefore, we need to understand aerosol dynamics, urban climate and turbulence, and to investigate all of them together as there are strong and highly complex, nonlinear interactions and feedbacks between these phenomena in different scales. Figure 1 illustrates a schematic picture on these interactions and feedbacks.

Fig. 1 Schematic picture showing the interactions and feedbacks between the different aerosol and meteorological processes. Acronyms: NPF—new particle formation, CCN—cloud condensation nuclei, UHI—urban heat island, BLD—boundary-layer dynamics. CCN can influence the energy/radiation balance directly or via aerosol–cloud interactions
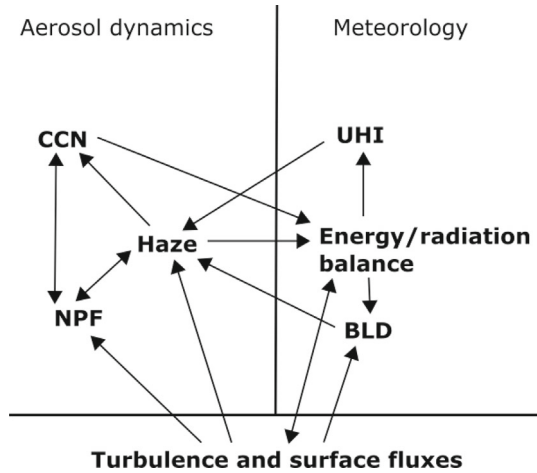
2.2 Turbulent Flow Over the Urban and Vegetation Canopies

Surface fluxes and atmospheric stability are strongly coupled via turbulent mixing, so that for example the aerodynamic coupling between the land surface and PBL influences the surface energy balance and is primarily controlled by the atmospheric stability. Typically, a negative feedback occurs during daytime when an increase in the surface temperature increases unstable atmospheric conditions and turbulent mixing, resulting in an increased sensible heat flux, which acts to reduce surface temperature. During stable conditions during nighttime, temperature profiles are inverted and turbulence is suppressed. The PBL height (BLH) represents the vertical extent of the atmosphere that is directly influenced by the Earth's surface. Therefore, the BLH has been used as a scaling parameter under a range of atmospheric stability conditions (Zilitinkevich et al. 2012) to characterize the exchange between the land surface and atmosphere.

In the lowest levels of the PBL, the atmospheric surface layer, momentum and heat/moisture transfer relationships are based on Monin–Obukhov Similarity Theory functions that depend on the static stability and scaling parameters, such as the roughness lengths for momentum and heat (Monin and Obukhov 1954). The roughness lengths are often taken as empirical values in models and assumed independent of flow characteristics. It is well known from laboratory experiments that while the transfer of momentum through the surface is mainly due to the pressure difference at the opposite facets of the roughness elements, the heat transfer is carried out by means of molecular heat conduction (Brutsaert 1982). Moreover, in contrast to the traditional assumption of a constant roughness length for momentum (z_0) fully characterized by the geometric properties of the surface, Zilitinkevich et al. (2008) theoretically derived and empirically demonstrated, that over very rough surfaces, including forest and urban canopies, z_0 depends on the stability parameter, and hence on the flow characteristics. Since the currently most commonly used roughness length formulations neglect this dependence, it results in strong and systematic overestimation of a surface resistance under stable stratification. The proposed formulation corrects for this drawback and can be implemented in meteorological simulations, largely to improve modelling of the most harmful air pollution episodes typical of very stable stratification in cities.

2.3 Turbulence Effect on New Particle Formation and Growth

Turbulence causes fluctuations in atmospheric concentrations of oxidants and precursors gases, such as ozone, OH, NO_3, Criegee intermediates and SO_2, CO, VOCs, as well as in temperature and humidity. All this together can enhance the rate of chemical reactions. For example, ozone formation is a highly nonlinear process depending mainly on concentrations of NOx, CO and VOCs, and fluctuations can either enhance or depress the ozone formation.

Atmospheric new particle formation is even a more nonlinear process than ozone chemistry, since particle formation rates typically depend on concentrations of low-volatile vapours, such as sulphuric acid, with the power-law function (typically to the power of 2–4). The survival probability of newly formed particles to CCN or accumulation (haze particle) sizes depends exponentially on the ratio between their growth rate (GR) and condensation sink (CS). The smaller is the value of CS/GR, the larger is the fraction of particles that will reach CCN or accumulation mode sizes (see Kerminen and Kulmala 2002; Kulmala et al. 2017). Since GR depends on the vapour concentrations and CS on the existing aerosol population, concentration fluctuations can significantly enhance or reduce atmospheric CCN production.

The effect of turbulence on heterogeneous nucleation has been investigated by Zapadinsky et al. (1995) using a theoretical model. The nucleation probability of water vapour onto an aerosol particle increases under conditions of the fluctuating saturation ratio around a certain mean value as compared to the case of non-fluctuating saturation ratio equal to this mean value. The magnitude of this enhancement depends on the saturation ratio (mean value and variance), contact parameter, and the ratio between the exposure time of the particle to the fluctuations and the characteristic correlation time for the saturation ratio, i.e. correlation time of turbulent eddies. This model can be generalized to organic vapours, and it can be expected that the nucleation probability of vapours onto aerosol particles is enhanced in a turbulent boundary layer. In addition, the particle velocity could be changed in the turbulent flow. The mixing process is accompanied by interactions between inertial particles and near-wall turbulent vertical structures (Li et al. 2016), which lead to settling and accumulation of the particles near the surface. This process will likely decrease the CS. In the regime with a smaller CS, a larger fraction of particles can survive to CCN sizes, which is another effect of turbulence favouring survival of the small particles, besides the influence of supersaturation fluctuations on the growth rate.

An interesting result was reported for the activation of cloud droplets under conditions of a fluctuating saturation ratio (Kulmala et al. 1997a, b). It was demonstrated that cloud droplets could be formed even at subcritical mean saturation ratio, provided that the variance is large enough. Another consequence of a stochastic saturation ratio field is a bimodality of the particle-size distribution, resulting from the difference of conditions for separate particles when some of them experience enhanced saturation ratio long enough to be activated while some do not.

In Fig. 2, we show which fraction of newly formed particles (3 nm) grow to CCN sizes (here 100 nm). Here, we assume that fluctuations can enhance the growth rate either by 20% or by the factor of 2. Even in the smaller enhancement, the survival probability is increasing significantly and the number of CCN is increasing. Particularly, this potential enhancement of growth rate is important in smallest particles (sub 10 nm) due to enhancing their survival probabilities. On the other hand, also the formation rate itself can increase even with significantly higher numbers. In the case of exponential dependence (power law with exponent 4), 20% enhancement of concentration of nucleating vapour will enhance formation rate at 1.5 nm by a factor of 2 and two times higher concentration—by a factor of 16.
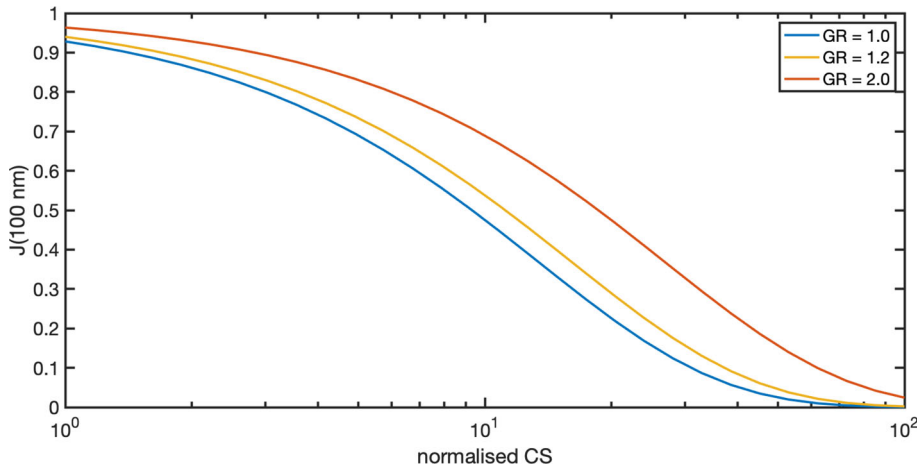
Fig. 2 The effect of turbulence/fluctuations on the fraction of particles surviving from NPF to CCN. The formation rate at 3 nm is $1.0 \; \mathrm{cm}^{-3} \; \mathrm{s}^{-1}$. CCN size in this simulation is 100 nm. The condensation sink is normalized to be one (when it is $10^{-4} \; \mathrm{s}^{-1}$). The used growth rates are $1.0 \; \mathrm{nm} \, \mathrm{h}^{-1}$, $1.2 \; \mathrm{nm} \, \mathrm{h}^{-1}$ and $2.0 \; \mathrm{nm} \, \mathrm{h}.^{-1}$

2.4 Turbulence and Continental Biosphere–Atmosphere Cloud Climate (COBACC) Feedback Loop

Climate-biosphere feedbacks involve phenomena of very different scale and origin. Albedo and evapotranspiration control near-surface budgets of energy and matter (surface fluxes) and, in turn, are essentially controlled by ecosystems—through partitioning of the surface energy budget between sensible and latent heat fluxes (Brunsell et al. 2011). In addition, local microclimate is controlled by physical processes in an atmospheric PBL: turbulence, radiation, evaporation, etc. The PBL is affected by local ecosystems, and it traps local impacts due to strongly reduced turbulent exchange at the PBL upper boundary. At local scales, a mosaic of microclimates, associated ecosystems and ecosystem responses is maintained, and this feeds back to the local climate (Muster et al. 2015; Davy and Esau 2016; Miles and Esau 2016; Davy et al. 2017).

The feedbacks between ecosystems and PBL are still insufficiently understood (Eugster et al. 2000; Pavlov and Moskalenko 2002; Blok et al. 2010; Barichivich et al. 2014; Ford and Frauenfeld 2016; Nauta et al. 2015). Quantification of these feedbacks is essential for understanding and modelling of interactions between ecosystems and climate (Jeong et al. 2011; Loranty et al. 2014). Adjustments of the PBL to large-scale features of climate depend on both global warming and surface fluxes. Here, the key is the height of PBL, determining its sensitivity to external impacts. The PBL height depends crucially on the properties of the underlying surface/terrain, including the height and structure of canopy (Miao et al 2015; Auvinen et al. 2020), leaf area index (LAI) (Adedipe et al. 2020), albedo and thermophysical properties of the soil (Lee and De Wekker 2016; Zhang et al. 2018; Xu et al. 2021). Capping inversions at the upper boundary of the PBL restrict the exchange of energy and matter between the PBL and free atmosphere, and to a large extent block local impacts of the surface within the PBL. The PBL height quantifies the largest possible length-scale of the PBL's own motions, integrating finer-scale impacts from the surface and determining microclimate's vertical and horizontal dimensions (Zilitinkevich et al. 2015).

To address feedbacks between ecosystems, PBL and climate, a Continental biosphere–atmosphere cloud climate (COBACC) feedback loop was proposed (Kulmala et al. 2014). It is a carbon-based loop resulting from the complex interactions between atmosphere and biosphere. The idea behind this feedback loop can be formulated as follows: the increase in the atmospheric CO_2 concentration increases ambient temperatures and stimulates photosynthesis. These changes lead to increased gross primary production (GPP) and stronger emissions biogenic volatile organic compounds (BVOC) into the atmosphere. BVOCs are precursors of low-volatile vapours responsible for the formation and growth of atmospheric aerosol particles. Subsequently, concentrations of secondary organic aerosol particles will be increased, enhancing the scattering of solar radiation back to space and increasing the diffuse fraction of total radiation, the latter of which enhances photosynthesis. The increase in GPP closes the loop, resulting in a positive feedback. In addition, aerosol particles can serve as CCN, via aerosol–cloud interactions, adds further complexity to the concept of the feedback loop (Artaxo et al. 2022).

With additional considerations, the COBACC feedback loop can be utilized in estimating the total radiative effect of ecosystems, including the carbon sink, albedo changes, and aerosol-radiation and aerosol–cloud interactions. This CarbonSink + concept have been utilized in boreal forest using the data from the SMEAR II station, Hyytiälä, Finland (Kulmala et al. 2020).

Turbulence in the boundary layer and residual layer enhances formation and growth of aerosol particles (e.g. Zapadinsky et al. 1995; Kulmala et al. 1997a, b; Wehner et al. 2010; Wu et al. 2021), increasing scattering effect (Ezhova et al. 2018). Vertical updrafts in the convective boundary layer provide vertical aerosol transport within the boundary layer and contribute to the cloud formation. In its turn, aerosol population can affect stability of the boundary layer. Su et al. (2018) reported the relationship between PBL height and PM for China: stronger for polluted sites, weaker for cleaner sites. Depending on the vertical distribution of aerosol, radiative effects can be localized in the lower, middle or upper boundary layer decreasing or increasing its stability (Su et al. 2020), and hence, influencing conditions for aerosol formation and growth.

Overall, since turbulence is typically enhancing aerosol production even crucially and also somewhat enhancing aerosol growth rates, turbulence is enhancing SOA formation and therefore also COBACC feedback loop and CarbonSink + . However, the exact values are hard to estimate and more investigations in different environments are needed.

3 Planetary Boundary-Layer Dynamics and Interactions

There have been numerous theoretical and experimental studies of boundary-layer dynamics already for over a century. Also, interactions between the haze and BLH have been the focus of many studies in recent decades. Therefore, the direct effect is well known where the aerosols attenuate the incoming solar radiation, which decreases the energy reaching the surface thereby decreasing the surface heat fluxes and turbulence. These, together decrease the BLH which forces the near-surface pollutants into a smaller volume, further enhancing the accumulation of pollutants (e.g. Baklanov et al. 2014; Petäjä et al. 2016). However, there are several processes, interactions and feedbacks between the boundary-layer dynamics, meteorological conditions and aerosol dynamics that are still poorly understood. Understanding the behaviour of the planetary boundary layer is crucial for understanding the influences

of climate change and for tackling air quality challenges. Only online, coupled meteorology–chemistry models, together with comprehensive observations, can provide a proper understanding of these phenomena, even though there are still many uncertainties associated with the use of these models (e.g. Zhang 2008; Baklanov et al. 2014).

In this chapter, we discuss the important two-way interactions between the PBL meteorology and air pollution and show two case studies: a modelling example on the direct and indirect effects of aerosols on the BLH in European cities, and a measurement example on effect of haze on both BLH and UHI in a highly polluted megacity in China.

3.1 Two-Way Interactions Between PBL Meteorology, Air Pollution and Aerosols

The interaction of atmospheric aerosol particles with the PBL is associated with many different mechanisms, in particular their influences on the radiative budget due to aerosol–radiation and aerosol–cloud interactions. Several examples of the loops/chains of interactions between aerosol particles, gases, PBL and components of the Earth system are demonstrated in Fig. 3.

In general, we can distinguish several processes and feedbacks between the PBL dynamics and air quality. It is important to notice that PBL meteorology—air quality interactions and feedbacks work in both directions. The effects of meteorology and climate on gases and aerosols can be summarized as follows:

– PBL meteorology is responsible for atmospheric transport and diffusion of pollutants;
– Changes in PBL turbulence, temperature, humidity and precipitation directly affect species concentrations;
– Changes in PBL vertical temperature and turbulence structure affect formation and transport of species;
– Changes in vegetation alter dry deposition and emission rates of biogenic species;
– Climate changes alter biological sources and sinks of radiatively active species.

The effects of primary and secondary aerosol particles on meteorology and climate, in turn, can be summarized as follows:
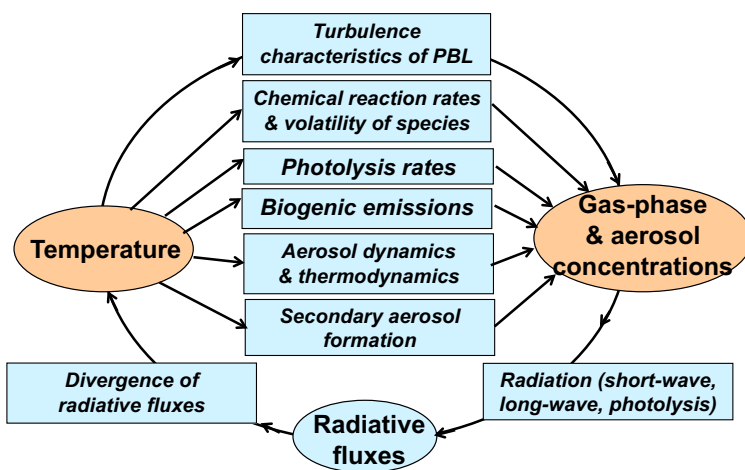

Fig. 3 Interactions loops/chains between aerosol particles, gases and PBL. Conceptual model of PBL impacts from temperature on concentrations and vice versa (modified after Baklanov et al. 2014)

– Decrease net downward solar/thermal-IR radiation and photolysis (direct effect);
– Affect PBL meteorology (decrease near-surface air temperature, wind speed, and cloud cover and increase RH and atmospheric stability) (semi-indirect effect);
– Aerosols serve as CCN and by that way increase droplet number concentrations, reflectivity and optical depths of low-level clouds (LLC) (the Twomey or first indirect effect);
– Aerosols increase liquid water content, fractional cloudiness and lifetime of LLC but suppress precipitation (the second indirect effect).

More detailed classifications of these effects, parameters and model variables have been presented in Baklanov et al. (2014, 2015). Concerning turbulence in the PBL, aerosol particles and associated interactions, we modified these classifications and summarized them in Table 1.

The interactions of aerosols with gas-phase chemistry and their impacts on radiation and cloud microphysics are strongly depending on their physical and chemical properties. There is still a strong need for research and improved understanding of these phenomena before these interactions could be incorporated into seamless meteorology–atmospheric composition forecast models (Baklanov et al. 2015).

As stressed by Baklanov et al. (2014), in order to better understand chains and loops of interactions, instead of only looking at the meteorological parameters affecting chemistry, it is also important mentioning the effect of altered meteorology on meteorology where altered meteorology can affect other meteorological variables/processes (Table 1). For example, clouds modulate boundary-layer outflow/inflow by changes in the radiative fluxes as well as alterations of vertical mixing and the water vapour modulates radiation. The temperature gradient influences cloud formation and controls turbulence intensity and the evolution of the PBL. Similar feedback mechanisms exist for altered chemistry impacts on chemistry. For example, biogenic emissions affect the concentrations of ozone and secondary organic aerosols. On a more general level, a number of chains and loops of interactions take place:

(a) A loop feedback starting with temperature that affects chemistry and thus chemical concentrations; the changes in chemical concentrations will in turn affect radiative processes (Table 1), which will then affect temperature to close the loop (illustrated in Fig. 3).
(b) A chain feedback starting with aerosol that affects radiation and thus photolysis and chemistry (Table 1).
(c) A chain feedback starting with temperature gradients that affects turbulence mixing (meteorology to meteorology feedback); thus affecting surface-level pollutant concentrations (Table 1) and boundary-layer outflow/inflow.
(d) A chain feedback starting with aerosols that affect cloud optical depth through influence of droplet number on mean droplet size (Table 1); the resulting changes in cloud formation will then affect the initiation of precipitation.
(e) A chain feedback starting with aerosol absorption of sunlight which results in changes in the temperature profile of the atmosphere and vertical mixing (Table 1) and thereby changes in the cloud droplet formation, which affects cloud liquid water and thus cloud optical depth.

Only online coupled meteorology–chemistry models permit correct descriptions of these two-way effects, chains and loops of feedbacks, although there are still many uncertainties associated with the use of these models (Zhang 2008; Baklanov et al. 2014, 2018).

Table 1 Meteorology and PBL interactions with atmospheric aerosols and chemistry

Meteorology's impacts on chemistry:

Wind speed and direction	Determines horizontal transport and vertical mixing of chemical species
	Influences dust emissions and sea-salt emissions
Atmospheric turbulence	Determines turbulent diffusion of trace gases and aerosols
	Affects formation and transformation of aerosols and other species
Planetary boundary-layer (PBL) parameters	Influence turbulent and convective mixing of gases and aerosols in PBL, intrusion from free troposphere, dry deposition at surface
PBL height	Influences concentrations, prevents transport of pollutants to free troposphere
Temperature	Modulates chemical reaction and photolytic rates
	Modulates biogenic emissions (isoprene, terpenes, dimethyl sulphide, etc.)
	Photo-synthetically active radiation: modulates biogenic emissions (isoprene, monoterpenes)
	Affects the volatility of chemical species
	Contributes significantly aerosol dynamics (coagulation, condensation, nucleation)
Temperature vertical gradients	Determines vertical diffusion intensity
Temperature and humidity	Affect aerosol thermodynamics (e.g. gas/particle partitioning, secondary aerosol formation)
Water vapour	Modulates OH radicals
Liquid water	Determines wet scavenging and atmospheric composition
Cloud processes	Affects mixing, transformation and scavenging of chemical compounds
Precipitation	Impacts on atmospheric composition
Soil moisture	Modulates dust emissions
	Influences dry deposition (biosphere and soil)
Surface-vegetation-atmosphere exchange processes	Affect natural emissions (e.g. dust, sea salt, pollen, nitrogen compounds, biogenic VOCs, CO_2, water vapour) and dry deposition
Lightning	Increases NOx emissions
Radiation	Modulates chemical reactions and photolytic rates
	Modulates biogenic emissions (isoprene, monoterpenes)

Chemical species' impacts on meteorology:

Aerosols	Modulate radiation transfers (SW scattering/absorption, LW absorption)

Table 1 (continued)

Chemical species' impacts on meteorology:

	Affect boundary-layer meteorology (temperature, humidity, wind speed, PBL height, stability)
	Extraordinary high concentrations can affect stability and wind speed
	Act as cloud condensation nuclei and ice nuclei
Aerosols physical properties (size distribution, mass and number concentrations, hygroscopicity)	Influences cloud droplet or crystal number and hence cloud optical depth and hence radiation
	Modulates cloud morphology (e.g. reflectance)
	Influence precipitation (initiation, intensity)
	Affects haze formation and atmospheric humidity
	Change scattering /absorption
Soot deposited on ice	Changes albedo
Radiatively active gases	Modulate radiation transfers

Altered meteorology impacts on meteorology:

Clouds	Modulate boundary layer outflow/inflow
Water vapour	Modulates radiation
Temperature gradient	Influence cloud formation
	Controls turbulence intensity

Altered chemistry impacts on chemistry

Biogenic emissions	Affect concentrations of ozone and secondary organic aerosols
Polymerization of organic aerosols	Yields long chain SOAs with lower volatility

3.2 Planetary Boundary-Layer Height and Air Pollution

The upper boundary of the PBL acts as a barrier preventing (or strongly reducing) vertical dispersion in most air quality and dispersion models, and essentially controls extreme weather events and microclimates in numerical weather prediction and climate models. Methods of calculation of the BLH have been the subject of numerous theoretical and experimental studies since Ekman (1905). Zilitinkevich, in his books Zilitinkevich (1970, 2013) and numerous scientific papers (e.g. Zilitinkevich 1972; Zilitinkevich and Mironov 1996; Zilitinkevich and Baklanov 2002; Zilitinkevich et al. 2002a,b, 2007a), substantially developed methods for the estimation of the PBL height in different regimes of stratified boundary layers as highlighted throughout this paper where these methods are discussed in more details.

In atmospheric chemical transport models, turbulent mixing takes place within the PBL and therefore the BLH is estimated at each time step in order to define up to which model level turbulence mixing exists. When a first-order closure is used, the height of the PBL is an essential parameter (implicit or explicit) in the description of mixing of meteorological properties (see overview in Seibert et al. 2000).

Atmospheric aerosol particles can influence the turbulent characteristics of PBL and in particular the BLH. Several studies have analysed different mechanisms of aerosol and PBL

interactions. Kong et al. (2015) conducted WRF-Chem sensitivity studies on Russian wildfires around Moscow in August 2010. Chains of aerosol direct and indirect effects on meteorology were analysed in that paper and concluded that there are:

- Significant aerosol direct effects on meteorology (and loop back on chemistry).
- Reduced downward short-wave radiation and surface temperature, and also reduced BLH. It in turn reduced photolysis rate for O_3.

The impact of aerosols on meteorology and BLH have been investigated in several studies and projects, including the COST Action EuMetChem (Baklanov 2017; Forkel et al. 2016) and AQMEII Phase 2 modelling exercise (Galmarini et al. 2015). In particular, Forkel et al. (2016) and Baró et al. (2017) analysed aerosol impacts on regional temperatures for cases studies covering two important atmospheric aerosol episodes over Europe in the year 2010: (i) a heat wave event and a forest fire episode (July–August 2010) and (ii) a more humid episode including a Saharan desert dust outbreak in October 2010. Makar et al. (2015) considered annual simulations with and without feedbacks on domains over North America and Europe for the year of 2010. The incorporation of feedbacks was found to result in systematic changes in the forecast predictions of meteorological variables, with the largest impacts found in summer and near large sources of pollution. Models considering only the aerosol direct effect predicted feedback-induced reductions in temperature, surface downward and upward short-wave radiation, precipitation and BLH, and increased upward short-wave radiation. The indirect effects included in the model were the same that are described in Sect. 3.1. The feedback response of models considering both the aerosol direct and indirect effects varied across models, suggesting that the details of implementation of the indirect effect have a large impact on model results, and hence should be a focus for future research. The feedback response of models incorporating both direct and indirect effects was also larger in magnitude to that of models with the direct effect alone, implying that the indirect effect may be the dominant process. In particular, impact of direct and indirect aerosol effects on the BLH [m] simulation for the European domain for the year 2010 using WRF-Chem model provided by Rahela Zabkar and Gabriele Curci is presented in Fig. 4 (Grid-average Feedback-Basecase).
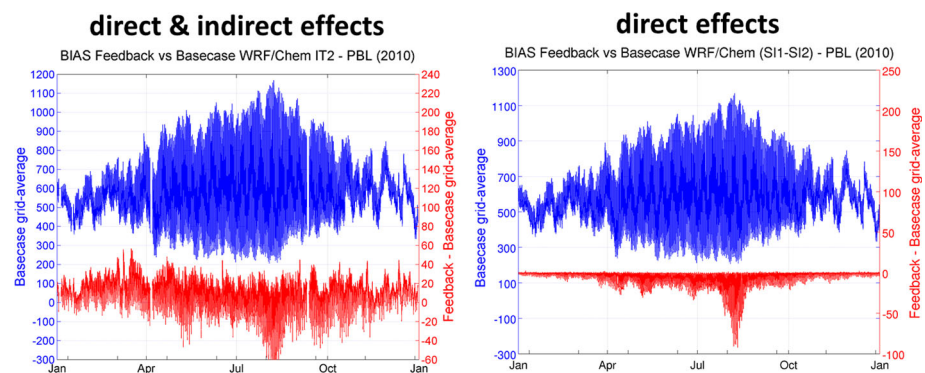

Fig. 4 Impact of direct and indirect aerosol effects on the BLH [m] simulation for the European domain for the year 2010 using WRF-Chem model (after Makar et al. 2015). The blue lines are showing the BLH from the Basecase simulation using WRF-Chem model without any aerosol effects included in the simulation. The redlines are showing the bias of BLH of the simulations including the direct and indirect aerosol effects (left panel) and only direct effects (right panel) when comparing to the Basecase (grid-average Feedback-Basecase)

When only direct effects were included in the simulations, the feedbacks always decreased BLH within the range [− 80: 0] metres, with the highest decrease in August (Fig. 4). However, when both direct and indirect effects were included, the feedbacks mainly increased the BLH in winter and during the summer the feedbacks both increased and decreased the BLH within the range [− 60: 40] metres (Fig. 4).

Within the WMO Working Group for Numerical Experimentations (WGNE) (Rémy et al. 2015), impacts of aerosols on meteorology were studied on example of an extreme dust storm event of 17 April 2012 over Egypt. This study demonstrated a nice example of the two-way interaction's chain/loop of dust aerosol and boundary-layer processes. Based on the results, the direct effect of aerosols increased the 10 m wind speed due to the increasing PBL inhomogeneity and forming stronger low-level jets, which leads into larger aerosol optical depths (AODs). A small increase in 10 m wind speed brings a large increase in dust aerosol production through saltation (power 3 dependency to 10 m wind speed). Correspondingly the increase in dusting intensity leads to increasing AOD and decreasing the solar radiation flux to the surface. The chain of interactions is closing into the loop. When including the indirect aerosol feedbacks, the uncertainties and the spread of the results were substantially increased between the different models used, which emphasizes that the indirect effects are not yet well accounted for in many models.

3.3 Urban Boundary Layer: Modelling the Effect of Heat Island and Aerosol

One of the most important and complex for modelling types of PBLs is the urban boundary layer (UBL). Specifics of UBL formation and development are characterized by the following aspects:

- Urban pollutants emission, their chemical transformation and atmospheric transport,
- Land-cover and land-use drastic changes due to urbanization,
- Anthropogenic heat fluxes and the presence of the urban heat island (UHI),
- Local-scale non-homogeneities, sharp changes of roughness and strong variability of heat fluxes depending on types of urban districts,
- Wind velocity reduction effect due to buildings of various heights,
- Redistribution of local eddies due to buildings, from large to small,
- Trapping of radiation in street canyons and shadowing effects,
- Effect of existing artificial surfaces—urban soil structure, differences in diffusivities heat and water vapour,
- Internal urban boundary layers, urban Mixing Height (MH),
- Effects of pollutants (especially, aerosols) on urban meteorology and climate, and especially, in urban and sub-urban areas,
- Urban effects on clouds, precipitation and thunderstorms.

For UBLs, due to their strong inhomogeneity and impacts of UHI, diagnostic methods for BLH are not completely suitable (see, e.g. Baklanov 2002). Therefore, in particular, for determination of the stable BLH for urban conditions it is suggested, as the best option for stable BLH estimation, to use a combination of the Zilitinkevich et al. (2002a) diagnostic parameterization together with a prognostic equation for the horizontal advection and diffusion terms (Zilitinkevich and Baklanov 2002). Implementation and testing of this approach into different meteorological models demonstrated better correlations with observations over urban areas compared with other commonly used methods (Baklanov et al. 2007).

In the FP7 EU MEGAPOLI project, Beekmann et al. (2015) and Baklanov et al. (2016) analysed UBL processes and interactions and tried to answer on the key question: What are

the key feedbacks between air quality, urban climate and global climate change relevant to megacities? As result, the following conclusions were achieved:

- The direct impact of climate change on air quality in megacities is significant due to temperature (BVOC fluxes, wildfires, deposition, O_3, CH_4, SOA, pSO_4, pNO_3), radiation (photolysis), cloudiness and precipitation changes.
- The coastal megacities climate change-induced increase in the temperature gradient between land and sea has resulted in more intensive and frequent sea breeze events and associated cooler air and fog.
- The impact of the direct aerosol effect was found to be substantial with regard to the turbulent characteristics of the airflow near the surface, as well as reduction of the BLH.
- The aerosol indirect effects can significantly modify meteorological parameters, such as daytime temperatures and BLH, while NO_2 concentrations are moderately affected.
- Compared to the direct and indirect aerosol feedbacks, UHI feedbacks can exhibit the same order of magnitude effects on MH, but with a strong sensitivity of chemistry and a strong nonlinearity.
- In general, UHI increases BLH, but in polluted cities the urban aerosols, due to direct effects, lead to decreasing BLH. The effects of urban aerosols on the BLH, could be of the same order of magnitude as the effects of UHI (i.e. about 100–200 m for stable boundary layer).

The DMI-Enviro-HIRLAM model simulations for a case study of 19 June 2005 for the Copenhagen (Denmark) and Malmo (Sweden) metropolitan areas showed (Fig. 5a) UHI impact on the BLH. Figure 5 shows the differences in the BLH, simulated for the same time by two versions of the DMI-Enviro-HIRLAM model. As seen in Fig. 5b, at low resolution (15 km) the model does not well represent the urban effects and hence, does not show a strong influence on the boundary-layer height over the Copenhagen and Malmo metropolitan area compared with the high-resolution urbanized run (Fig. 5a). The UHI effect, considered by the urbanized version, on the BLH over Copenhagen and Malmö (on the Swedish side) is more visible in Fig. 5a. The BLH, which is also the mixing height for air pollutants, effected considerably the air concentration and deposition levels of the contaminants.
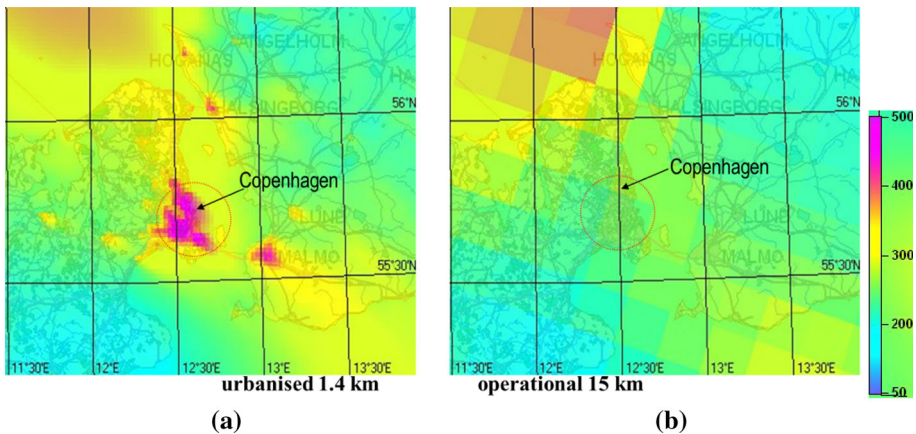

Fig. 5 Effects of the model resolution and urbanization (UHI) on the nocturnal boundary-layer height (in metres) on example of the case study—DMI-Enviro-HIRLAM urbanized runs at **a** 1.4 km and **b** non-urbanized 15 km resolution

The Enviro-HIRLAM model simulations considering different effects—aerosol, urban and both combined—for the MEGAPOLI Paris campaign (France, July 2009) in focus concluded the following results (Beekmann et al. 2015; Baklanov et al. 2016). For the air temperature at 2 m, the modelling results of Enviro-HIRLAM showed high positive correlation coefficients (> 0.8) against the observations. On a diurnal cycle, large variability was observed during the daytime compared with nighttime. When only aerosol or urban effects were accounted for in the simulations, the biases were in the similar range but opposite (aerosol effect negative in daytime and positive in nighttime, urban effect positive in daytime and negative in nighttime). When both, aerosol and urban effects were included, the biases in the simulations during the nighttime were reduced. Implementation of both aerosol and urban effects in meteorological models (especially, in operational weather forecasting) is important step towards improved numerical weather prediction, chemical weather forecasting and climate simulations (Shen et al. 2017; Solanki et al. 2022).

3.4 Urban Boundary Layer: Measuring the Effect of Urban Heat Island and Air Pollution

Haze formation, together with atmospheric boundary-layer processes, forms a feedback mechanism. Under weak synoptic forcing and clear sky conditions, haze attenuates the incoming solar radiation, reducing the surface energy availability and heat fluxes (Gao et al. 2020; Kajino et al. 2017). This decreases near-surface air temperatures (Ding et al. 2013; Wang et al. 2014), and furthermore the vertical turbulent mixing of air and the daytime BLH (Ding et al. 2016; Liu et al. 2018; Petäjä et al. 2016; Wang et al. 2018). At the same time, solar heat stored in urban surfaces during the daytime is reduced, weakening nocturnal heat emissions and urban heat island, and therefore also the nighttime BLH. The reduced BLH constrains diverse emissions into a smaller and smaller air volume, further enhancing accumulation of pollutants during haze episodes. Additionally, calm synoptic-scale winds enable UHI-induced circulations, also called the urban heat dome flow, which drives the transport of pollutants within urban regions (Fan et al. 2017b; Zhang et al. 2014). This circulation, together with a low-level entrainment flow of cleaner air from the rural areas, increases ventilation, which improves urban air quality (Fan et al. 2017a). Unfortunately, weak synoptic-scale winds that enable UHI circulation, are favourable also for haze formation (Zhong et al. 2018), which is decreasing the magnitude of UHI, thus restricting UHI circulation and ventilation of urban areas. The influence of haze on BLH and UHI varies between the seasons, and there are also indirect aerosol effects that might have influences opposite to those described above.

We examined the effect of haze on BLH and UHI in Beijing in 2018. The analyses included only the days without precipitation and substantial cloud cover. The UHI magnitude was calculated as the difference between the temperature observations at BUCT-AHL station in the Haidian urban district next to 3rd ring road (Liu et al. 2020) and the closest rural station south of Beijing (Daxing measurement station, 39.65° N, 116.70° E). Therefore, the UHI effect might not be representative for the whole Beijing, but it was assumed to represent the UHI effect of the observation station area when compared to the nearest rural area.

The decrease in the daytime BLH due to haze was the greatest in spring (929 m, 48%; Fig. 6d) and autumn (885 m, 56%; Fig. 6h), when also the BLH in clean conditions was the greatest. The influence of haze on the BLH was smaller in winter (579 m, 40%; Fig. 6b) and especially in summer (359 m, 25%; Fig. 6f).

The effect of haze on the nocturnal magnitude of UHI was clearly the highest in winter (1.9 K, 28%; Fig. 6a), whereas in spring (Fig. 6c) and autumn (Fig. 6g) the effect was small
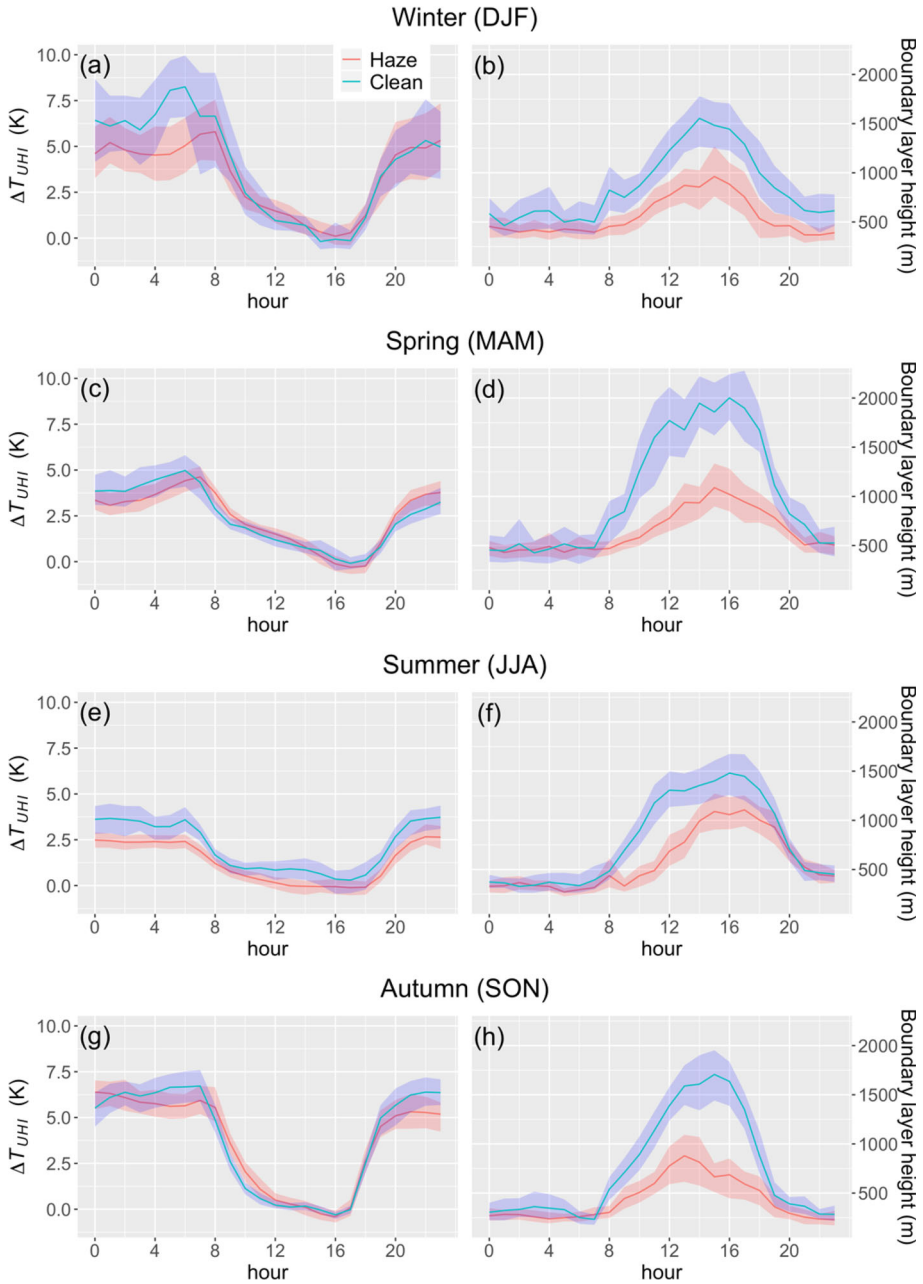
Fig. 6 The mean diurnal profile of urban heat island magnitude (ΔT_{UHI}, left column) and boundary-layer height (right column) for different seasons in 2018: winter (December, January, February), Spring (March, April, May), summer (June, July, August) and Autumn (September, October, November) for haze days (red line) and clean days (blue line). The shaded areas are the 95% confidence boundaries

(0.6 K, 13% and 0.4 K, 6%, respectively). The relative difference was the highest (31%) in summer (Fig. 6e), even though the absolute change was small (1.1 K). In winter, the nocturnal BLH was also clearly decreased by haze (126 m, 23%; Fig. 6b) due to a lower UHI magnitude by 28% (i.e. less heat available on surface) because of the attenuation of daytime incoming solar radiation by the haze.

3.5 Steps Towards a Roadmap for Future Activities

Planetary boundary layers (PBLs) have traditionally been distinguished by a single indicator, the static stability at the Earth surface and, accordingly, they have been classified as the three types (with typical occurrence time in parenthesis) (Oke 1988; Liu and Liang 2010): (nocturnal) stable, (daytime) unstable and (intermediate) neutral. In doing so, it is factually implied that the types are alternating in the 24-h circle, so that the lifetime of each type is about 8 h.

The conventional paradigm of PBL disregards a stable stratification, which the general circulation is slowly but persistently re-establishing in the free troposphere. This paradigm is factually based on the unspoken assumption that the lifetime of any of the three types of PBL is so short that the above slow mechanism of general circulation does not have time to manifest itself. Therefore, it is generally assumed that the static stability inherent in PBL is fully controlled by the sign of the vertical turbulent flux of heat at the surface: nocturnal negative (cooling), daytime positive (warming) and intermediate zero.

Historically, the vision on PBL stratification originated from an empirical knowledge on PBLs inherent in mid-latitudes over land, where most of the humankind lives, most of meteorological observations are carried out, and the 24-h diurnal cycle discussed above is guaranteed. It is not surprising that the conventional paradigm has received not only empirical but also psychological support. This is just the reason why the widespread long-lived stratification of PBLs maintaining the type of stratification (stable or unstable) much longer than 10 h were unnoticed for so long. Their principal difference from the familiar short-lived stratification of PBL is still neglected in boundary-layer meteorology and in operational weather, climate and air quality modelling.

In reality, long-lived stratification of PBLs is globally dominant (Zilitinkevich, 2013). The clearest examples are long-lived stable and unstable PBLs inherent to polar winter and summer, respectively, and lasting for up to several weeks. Another example, is long-lived unstable PBL typical for big cities due to the permanent anthropogenic warming of urban canopy (Dupont et al. 1999; Masson et al. 2008; Yu et al. 2013; Podstawczynska 2016). Such PBLs last long enough to be fully exposed to the impact of general circulation that, therefore, forms a permanent stable stratification in the upper part of PBL, thus reducing turbulence and making the PBL essentially shallower.

Now the concept of long-lived stratification of PBLs is only generally outlined (Zilitinke-vich and Calanca 2000; Zilitinkevich 2002; Zilitinkevich and Esau 2003, 2005; Zilitinkevich et al. 2007a; Petäjä et al. 2016; Ding et al. 2016). It is crucial to develop it further and apply to the mega-urban unstable PBL accounting for its following specific features:

- Very heterogeneous man-made landscape with various multi-scale constructions, forming a complex of wind patterns and very wide spectrum of turbulence
- Very high level of anthropogenic pollution
- Extreme anthropogenic warming over wide territory, causing in summer long periods of permanently convective urban PBL strongly affected by the imposed and, hence, comparatively shallow and sensitive to industrial emissions and heat waves.

To put the PBL studies into a wider applied research context, we define the Terrestrial Atmosphere-biosphere Interaction Layer (TAIL) as a system comprised of land surface (its topography, soils, inland waters, vegetation, man-made constructions, agriculture and industries) interconnected with an overlying essentially turbulent atmospheric PBL and weakly turbulent free atmosphere aloft. The PBL top acts as a "lid", restricting the impacts of the Earth surface to within the PBL and, thus, forming geophysical objects that can be called "PBL climates" (Zilitinkevich et al. 2015; Oke 1988; Oke et al. 2017). The lid traps the effects of warming or cooling and natural or anthropogenic emissions over hours to days, thus shaping microclimates, zones/cases of heavy air pollution, and many other specific or extreme features of climate or weather events that influence humans. The PBL top is also home to shallow clouds in about half of the continental TAIL area (Hagihara et al. 2010; Park and Shin 2019); clouds impact microclimates by modulating radiative fluxes. Terrestrial shallow clouds are coupled with fast PBL dynamics and exchanges of heat, mass (including particulate matter), energy and momentum at the surface, which makes them especially difficult to describe. Starting from the well-established premise that PBLs are "capped", we proceed further to propose the interdisciplinary concept of "primitive TAIL units" associated with clusters of a particular type of landscape (e.g. forest, wetland, residential neighbourhood) of order 1 to 10 square kilometres (typical areas subjected to the PBL's own mixing). We postulate that local, mesoscale, regional and global TAIL systems are composed of primitive TAIL units that share analogues to living organisms composed of cells. The TAIL regime is crucial for our planet and humankind. Actually, our future depends significantly on the progress how we understand processes at different scales and their interactions and feedbacks, also described in Sects. 2 and 3.

In spite of some progress, there are still several open questions, including:

- How turbulence will affect atmospheric chemistry and aerosol dynamics, particularly secondary aerosol formation
- How much aerosols influence turbulence and under which concentrations and characteristics of aerosols, and is the influence nonlinear?
- What are the effects of boundary-layer dynamics on haze formation and the effect of haze on boundary-layer dynamics?
- How turbulence is affecting on feedback mechanisms and interactions between earth surface and atmosphere, e.g. COBACC feedback loop? And how different feedbacks will affect turbulence?
- How urban heat and pollution islands are interacting and affecting turbulence?
- How pollution will reduce/enhance turbulence? Is there a local maximum if we look the turbulent transport as a function of pollution?

All these questions are related to mass and heat flows inside the PBL. Therefore, understanding these processes and answering the open questions are crucial in order to meet environmental grand challenges, particularly climate change, air pollution, and water and food supply. We need to utilize several tools, such as: (i) continuous and comprehensive in situ observations in different types of environments or ecosystems and platforms, (ii) ground- and satellite-based remote sensing and (iii) multi-scale modelling efforts, in order to provide improved conceptual understanding over the relevant spatial and temporal scales. When needed, we can utilize targeted laboratory experiments from different rigorous experiments like CLOUD experiments (e.g. Kirkby et al. 2011) at CERN and laboratory experiments performed at TROPOS (Leipzig, Germany, e.g. Sipilä et al. 2010).

One of tools to be used is science diplomacy (Royal Society, 2010; Lappalainen et al. 2022a,b). An example of this is the Pan-Eurasian Experiment (PEEX). The Pan-Eurasian

continent and particularly boreal and arctic areas in the PEEX territory are crucial for understanding future climate and also air quality–climate interactions (Kulmala et al. 2015; Lappalainen et al. 2016, 2022a,b). Therefore, the key to answering our research questions is to ensure the further development of continuous comprehensive observations in the PEEX geographical domain and the associated data flows (Hari et al. 2016; Alekseychik et al. 2016; Vihma et al. 2019; Petäjä et al. 2021). The existing PEEX international collaboration consists of more than 80 Universities and research institutes (Kulmala et al. 2015). Within the PEEX collaboration, we will focus, e.g. on quantification of COBACC loop and its different steps, like (a) the effectiveness of changing ecosystems to act as carbon sink, (b) present and future BVOC emissions and concentrations, NPF and GTP in boreal area, and the effect of changing aerosol load on carbon sink via increased levels of diffuse solar radiation as well as on CarbonSink + .

In future, we need to solve in quantitative manner at least following wide topics: (a) how BLH is connected with aerosols and other air pollutants?, (b) how UHI and air pollution are interacting?, (c) how turbulence will affect aerosol nanophysics and atmospheric chemistry?, (d) what is the effect of boundary-layer dynamics on COBACC feedback loop generally and NPF particularly?, (e) how changing climate will change boundary-layer dynamics?, (f) how important are different BL meteorology—aerosol feedback loops/chains and what are the requirements for seamless coupled chemistry-meteorology models (CCMM)?

Generally speaking, one of the big ideas from Sergej Zilitinkevich was to combine boundary-layer meteorology with atmospheric chemistry and aerosol dynamics. Therefore, he started developing the TAIL concept. Together with a global network of comprehensive, long-term observations producing open data, Zilitinkevich's theoretical understanding, combined with a deep understanding of aerosol nanophysics and atmospheric chemistry, can provide enough knowledge to solve global grand challenges. As summarized above, the role of PBL has been recognized as a critical field of science to society due to its direct implications on weather, air quality and climate, and furthermore, e.g. on agriculture and forestry. A deep understanding of PBL processes is needed for improved weather, air quality and climate models and predictions to support decision making. Thus, a specific international approach, a roadmap, is needed to address the role of PBL in atmospheric sciences but also a study area relevant for the environmentally safe society. Only this way, it is foreseen that within a decade we will have proper answers to present open questions.

The roadmap would help us to make a full use of existing data and to plan and carry out new observations. Thus the linkages to Sustainable Arctic Observation networks (SAON), World Meteorological Organization (WMO)—The Global Atmosphere Watch (GAW) Programme, GlobalSMEAR (Stations for Measuring the Earth Surface and Atmospheric Relations) and other observation networks should be connected to PBL dedicated approach. The roadmap should also be built on existing research programmes like Pan-Eurasian Experiment Programme (PEEX) or research networks like AASCO—Arena gap analysis of the existing Arctic science co-operations. Recent regional analysis of the future research needs should be integrated to the PBL approach (Petäjä et al. 2016; Lappalainen et al. 2022a).

At the same time, joint international research projects, especially together with Russia and China, would act as important science diplomacy asset. Mankind has roughly a 30-year time window to find joint understanding and practical solutions for the Global Challenges, such as climate change, sufficiency of the food supplies and energy (Kulmala et al. 2016). Based on the current trends, the atmospheric CO_2 concentrations will exceed value of 500 ppm in year 2050. The different political decisions of Russia and China are and will be affecting the atmospheric composition in their regions but will also having a global scale impact on the future development of mankind. Joint research forces are needed to ensure that the

research outcome is used as the baseline for policy actions, especially climate and air quality legislation, having impacts over country boarders and over a wide range of geographical areas. Joint research approach would also enhance trust between partners and facilitate that way also the route towards open data.

The roadmap includes: (I) Definition of open questions; (II) Finding out needed tools; (III) Description of research programme; (IV) Finding out proper funding organizations; (V) Description of projects; (VI) Implementation of the research programme; (VII) Collecting and obtaining open data; (VIII) Scientific results; (IX) Outreach; (X) Evaluation of Impact.

In this paper, we have already outlined several open questions and also discussed the needed tools. However, it is important to develop these further. The Phases III and IV should take place in 2022–2023 via several seminars and workshops and Phase V in 2023 via several open calls. The programme itself should be 2024–2032 (Phases VI–VIII) and all the time outreach (Phase IX) is needed. The evaluation of impact (Phase X) can start earlier than at the end of project. However, the final impact can be seen after a decade or even decades.

Data Availability The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

Declarations

Conflict of interests The authors have no competing interests to declare that are relevant to the content of this article.

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