UNIVERSITY OF PÉCS

Doctoral School of Earth Science

Numerical simulation of the fog

Ph.D. Thesis

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1 Introduction:

Fog forms, strengthen, and disperses due to complex interactions among diverse local, microphysical, dynamical, radiative, chemical processes, and boundary-layer conditions (Gultepe et al., 2007). Aerosol particles play a vital role in fog formation, development, and dissipation. Furthermore, the fog can also impact aerosol particles' characteristics (e.g., size distribution, chemical composition). The primary sources of particle numbers in cities are traffic, thermal power stations, factories and household emissions, and their concentration is affected by advection and scavenging processes. While scavenging by activation strongly depends on the chemical composition of the particles (e.g., Gilardoni et al., 2014), the collision scavenges of particles is mainly affected by size of the aerosol particles and that of the water drops.

The chemical composition and the size distribution of aerosol particles are affected by their sources, gas-phase chemical reactions and microphysical processes, and chemical reactions that occur in the water drops. Dissolution of some ambient gases into droplets and the subsequent aqueous-phase chemical reactions can also modify the particle size (Kerminen and Wexler, 1995; Meng and Seinfeld, 1994). Besides, considering the chemistry in the numerical simulation of fog is a great challenge due to the strong interaction between the different chemical processes and due to the lack of observational data of trace gases and various relevant inorganic and organic compounds for validating the models.

In this study a recently developed bin scheme (Schmeller and Geresdi, 2017) was used in a one – dimensional (1D) model to investigate, how the environmental conditions affect the physical and chemical characteristics of the fog. Although the 1D models cannot simulate some crucial characteristics of the fog (e.g., turbulence, radiative cooling, sedimentation), they allow for the very accurate simulation of specified processes such as microphysical and chemical processes. For example, Xue et al. (2019) carried out 1D model experiments using a bulk liquid chemistry scheme incorporating detailed SO_2 oxidation chemistry to derive SO_4^{2-} production over the full range of SO_2 atmospheric concentrations. Furthermore, rigorous model studies have been made to improve fog forecasting over this region (Pithani et al., 2020, 2019a, 2018). However, there is considerable scope to improve forecasting and detailed understanding of fog microphysics and chemical properties during the fog life cycle.

2 Research Objective

Budapest and Delhi are the capital cities of Hungary and India, respectively. These cities have different characteristics based on population density, traffic, industrialization, and agricultural activities. In Hungary, fog is also a frequent phenomenon during the winter. The interaction between air pollution and fog can also result in a weather situation that can cause a severe emergency or economic damage. Such is the drastic reduction in visibility, the formation of dense fog, smoke in the cities, which can, among other things, make aviation impossible and causes severe accidents in road transport. Since fog has a very high damage potential (physical and economic), it is necessary to develop objective tools for forecasting the characteristics, such as the timing of onset, intensity, and duration of fog, well in advance. For better fog forecasting, it is not enough to understand large-scale to mesoscale, micro-meteorological processes, but it is essential to know more about the microphysical processes that occur in fog to improve the parameterization of these processes.

Main aims of this research:

- Understand the impact of hygroscopicity and size distribution of aerosol particles on fog formation.
- Understand the impact of the scavenging process in the washout of the aerosol particles during the fog event.
- Understand the impact of the chemical process in the fog droplets on changing the size and hygroscopicity of the aerosol particles.
- Evaluate the Weather Research and Forecast (WRF) model performance in fog prediction.

3 Research Methods

3.1 1D model experiment

3.1.1 Observations:

For the Budapest case, T, RH, the size number distribution and hygroscopicity of the aerosol particles were obtained in a field project at the Budapest platform for Aerosol Research and Training (BpART) Lab (Salma et al., 2016) in Budapest from 9 December 2014 to 9 February 2015 (Enroth et al., 2018). Concentrations of SO_2, NH_3, HNO_3, O_3 provided by HMS are used to give the trace gas concentration in the atmosphere for the Budapest case. Less-hygroscopic (LH) and nearly hydrophobic (NH) particles are separated based on the measurements of the hygroscopic growth factors (HGFs) at different dry diameters of 50, 70, 110 and 145 nm. Measured hygroscopicity parameters of less-hygroscopic (LH) and nearly hydrophobic (NH) particles were 0.2 and 0.03, respectively.

For the Delhi case, we use the data observed in intensive observational periods during winter fog experiments (WIFEX; Ghude et al., 2017). To validate the model, we obtained T, RH, and number size distribution of dry aerosol particles in a diameter range of 10–300 nm (Ghude et al., 2017; Pithani et al., 2020). Trace gas concentrations of SO_2, NH_3, HNO_3 were measured and published by Acharja et al.(2020) for the exact location, but a different fog event was given as initial conditions due to the lack of available trace gas concentration data on 30 December 2016. The measured mean hygroscopicity parameter is 0.147, and less-hygroscopic (LH) and nearly hydrophobic (NH) particles 0.2 and 0.03 are derived from mean hygroscopicity, respectively (detailed equation mentioned in Jeevan Kumar et al., 2021).

3.1.2 Model description

A detailed bin scheme with moving boundaries was used to simulate the diffusional growth or evaporation of water drops, as published by Geresdi and Rasmussen (2005). The model involves 28 bin categories in the Budapest case and 95 bins in the Delhi case (the number of the bins depends on the size resolution of the observation) for both the hygroscopic and hydrophobic particles.

The numerical model involves the following processes:

- Model mimicked by the cooling rate, and the warming rate is tuned to fit the time profile of the simulated temperature to the observed one. The fog formation is simulated by holding the water content (sum of vapour and liquid) constant and gradually decreasing the temperature.
- The liquid water content in each bin is evaluated by solving the diffusional growth equation. The integration is performed in a time step of 0.0001 s. This small-time step allows us to avoid overestimating the diffusional growth in the case of tiny drops when the solution is dense. Furthermore, the competition for available vapour among drops with

different sizes and with different types and sizes of aerosol particles inside them can be taken correctly into consideration.

- The scavenging of aerosol particles (r <1.0 μm) by water drops through Brownian motion and phoretic forces. Theory published by Pruppacher and Klett (2010) is used to evaluate the impact of Brownian and phoretic impaction scavenging. While Brownian scavenging is efficient if the particle size is less than 0.1 μm, phoretic scavenging can reduce the concentration of the particles with a radius of about 0.1 μm (Santachiara et al., 2013). The latter process persists only in a subsaturated environment so that it can happen during either the formation or dissipation of the fog. In this study, we focus on the scavenging efficiency of Brownian motion and phoretic force. The impacts of gravitational collection and turbulence are not taken into consideration. The scavenging of particles with a radius less than 1 μm, both hydrophobic particles and haze, due to the Brownian motion and phoretic scavenging are calculated separately.
- The absorption and desorption of the trace gases of SO_2, NH_3, H_2O_2, HNO_3, O_3 and CO_2. Sulfate formation in the liquid phase due to oxidation of S(IV) by O_3 and H_2O_2. The amount of the dissolved trace gases and that of sulfate formed due to the chemical reaction were evaluated for each bin with a droplet radius larger than 1 μm. The stiff, ordinary differential equations about the chemical processes are solved by using time step of 0.0001 s.

Total 20 sensitivity experiments had been done to understand the impact of trace gas concentrations, aerosol hygroscopicity and their size distribution on the fog microphysics and chemistry. Among these 20 experiments, 12 tests belong to Budapest case (BLK1 – BMK4) and 8 tests belong to Delhi case (DSK1 – DMK1). Further detailed experiments mentioned in Table 1 in Jeevan Kumar et al., 2021.

3.2 WRF model experiment

3.2.1 Observations

For this modeling experiment we have taken the fog event occurred on 24-November-2020 morning time as a case study. The diurnal cycle of temperature, relative humidity, visibility, and wind speed were observed at Pecs and Szeged meteorological stations.

3.2.2 Model description

Several sensitivity experiments have been accomplished to study how the accuracy of fog forecast depends on planetary boundary layer (PBL) schemes implemented in the WRF Advanced Research Core (ARW, V4.3). The horizontal extension of the model domain is 800 km × 740 km with horizontal spatial resolution of 2 km.

Meteorological initial and boundary conditions are provided by reanalysis products of the European Centre for Medium term Weather Forecasting (ECMWF). ERA5 data are operational global analysis available on 0.25°×0.25° grids with 1 h temporal resolution.

The following parameterization schemes are set to simulate the different physical processes:

(i) The rapid radiative transfer model (RRMTG) for both longwave radiation and shortwave radiation (Iacono et al., 2008);

(ii) The Thompson aerosol aware two-moment bulk scheme for microphysics (Thompson and Eidhammer, 2014),

(iii) Noah land surface scheme to simulate the impact of the soil and land use (Chen and Dudhia, 2001).

(iv) Four different planetary boundary layer schemes used for sensitivity experiment which are, MYNN3.0 (Nakanishi and Niino, 2006), YSU (Hong et al., 2006), QNSE (Sukoriansky et al., 2005) and ACM2 (Pleim, 2007a).

4 Results and conclusions

4.1 Impact of Scavenging

Sensitivity experiments was made to investigate the scavenging efficiency of Brownian motion and phoretic force on the sub-micron particles in the fog. The detailed assessments located in Section 6.2 in the dissertation. The sensitivity experiments showed the following:

- The impact of the Brownian motion is dominant in all the investigated cases (Figure 1), the role of the phoretic scavenging mechanism is negligible in the Budapest cases (moderate polluted condition) and is more significant in the Delhi cases (high polluted condition).
- Due to the low supersaturation, particle scavenging through activation nucleation is less important than other mechanisms, such as Brownian and phoretic. On an average, 40–50% total particles are collected due to impact scavenging (Figure 1), and less than 1% of the LH particles are activated due to the low supersaturation and small hygroscopicity of the LH particles (refer to Table 2 in Jeevan Kumar et al., 2021).
- Sensitivity experimetns showed liquid chemical processes have no impact on the scavenging processes in either the Budapest or the Delhi cases (Figure 1).
- The larger reduction (ca. 25%) of particle concentration in the Delhi case proves that the efficiency of Brownian scavenging is higher if the small aerosol particles (0.1μm) are mostly NH particles (Figure 2 & Figure 3).

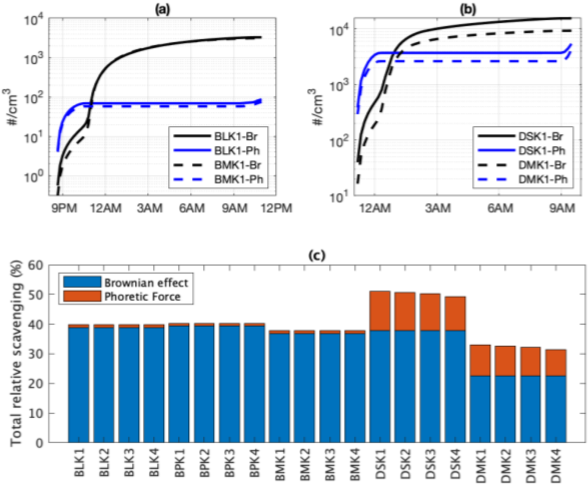

Figure 1: Cumulative effect of Brownian and phoretic forces on scavenging process in (a): Budapest case and (b): Delhi case. (c): Efficiency of Brownian motion and that of phoretic forces in total scavenging rate. The total relative scavenging means the ratio of the scavenged particle concentration and the total initial aerosol concentration.

Sensitivity experiments revel the time evolution of size distribution of dry aerosol/aerosol inside the droplets over the Budapest and Delhi cases, as well as the size distribution of droplets.

- During the onset period of the fog, the significant decrease of aerosol concentration at the radius smaller than 0.05 μm is the consequence of Brownian scavenging both in the Budapest and Delhi case.

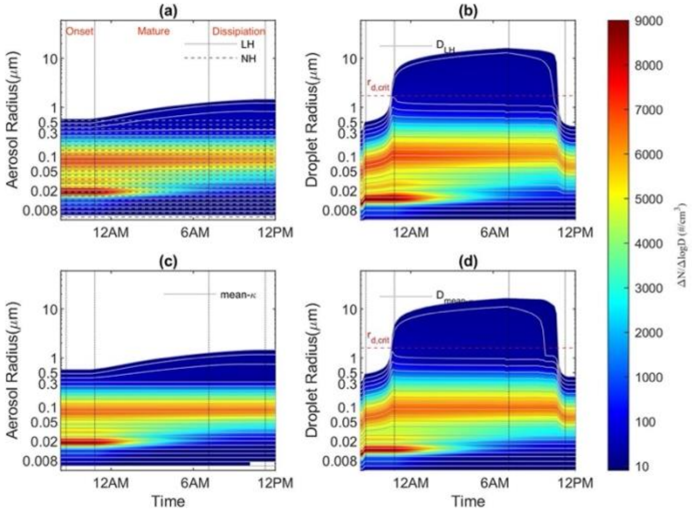

Figure 2: Contour plots depict the time evolution of aerosol and haze/droplet size distribution for Budapest cases (BLK1 at first row and BMK1 second row). The vertical dotted lines denote the different phases of the fog, such as onset, mature, and dissipiation (see the vertical dashed lines in Fig. 4 in Jeevan Kumar et al., 2021). r_{crit} means the critical radius for water drops at the supersaturation which occurs during the mature phase of the fog, LH and NH mean less hygroscopic and nearly hydrophobic dry aerosol particles, respectively. Mean-κ means that all dry aerosol particles have the same hygroscopicity, D_{LH} denotes drops formed on LH particles. $D_{mean\text{-}\kappa}$ denotes drops formed on aerosol particles having the same hygroscopicity. The time evolution of the bin boundaries (horizontal lines) for the aerosol particles and liquid drops are depicted as it is given by the figure legend.

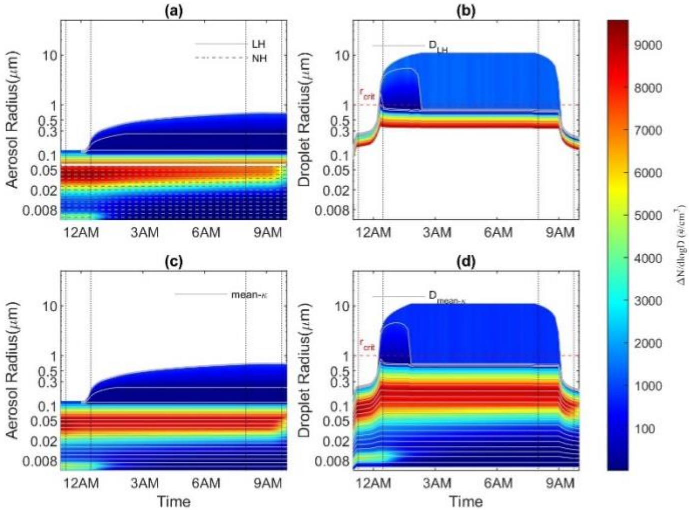

Figure 3: Contour plots depict the time evolution of aerosol and haze/droplet size distribution for Delhi cases (DSK1 at first row and DMK1 second row; refer to Figure 2).

- The concentration of aerosol particles at the size of about 0.1 μm is significantly larger in the Delhi case, than in the Budapest case (Figure 2). At this aerosol size, phoretic scavenging is more dominant than Brownian scavenging, because the Brownian effect is small if the aerosol size is near or larger than 0.1 μm.

- During the onset and dissipation periods, aerosol particles with a radius around 0.1 μm can only be captured by water drops due to phoretic forces in a subsaturated environment. The smaller warming rate and the larger hygroscopicity of water-soluble aerosol particles resulted in longer dissipation and subsaturated periods in the Delhi case.

- In the mature phase of the fog, drops larger than the critical size keeps growing, and they collect the sub-micron particles of the LH and NH modes through Brownian motion (Figure 2a & Figure 3a).
- Drops smaller than the critical size evaporate slightly even during the mature phase of the fog (see the time profiles of the bin boundaries for the drops/haze particles in Figure 2 & Figure 3).
- Comparison of the size distribution of aerosol particles in the onset and the dissipation period reveals the impact of the scavenging process promots the sulphate formation occurring inside of the drops.

4.2 Impact of liquid chemistry

The initial composition of a droplet is determined by the dissolution of soluble materials contained within an aerosol particle, which serves as the cloud condensation nucleus (CCN). Further variations in the composition come from subsequent scavenging of other non-activated, interstitial particles and from uptake of water-soluble gases and aqueous-phase reactions. The sensitivity experiments in the fog liquid chemistry showed the following:

- The acidity of the drops is affected not only by the liquid chemistry, but it also depends on the diffusional growth of the drops. While the increase of the LWC results in the increase of the bulk pH, the evaporation of the drops in the dissipation periods significantly reduces the pH (Figure 4g, h).
- Due to the long duration of the fog, the droplets absorb significant amounts of different trace gases (Figure 4). Since the presence and absorption of ammonia promote the absorption of SO_2 the sulphate formation is very efficient in the simulated cases (Figure 4i,j).

- S(VI) formation is highly depend on the availability of SO_2 in the environment and availability of HNO_3 in the environment is increase the quantity of formation of NH^+_4 along with NO_3^-.

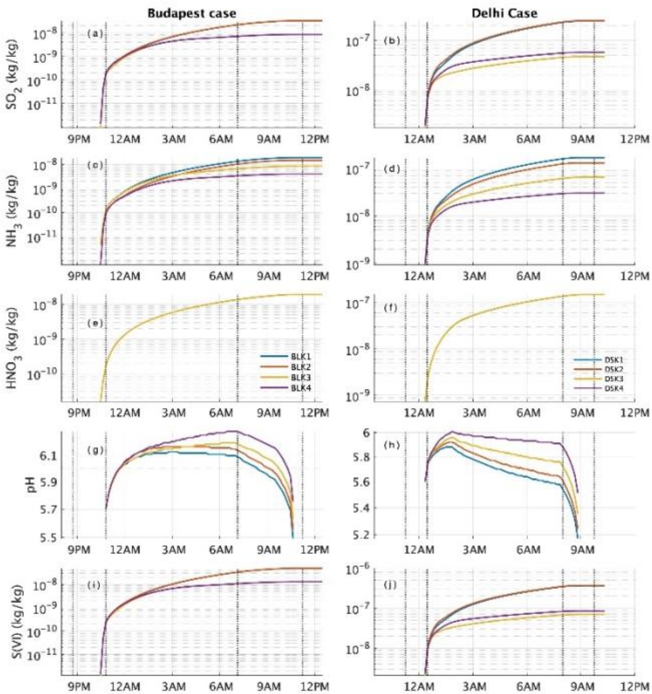

Figure 4: Time evolution of the accumulated absorption of trace gases (panels (a) – (f)), bulk pH (panels (g) and (h)) and accumulated S(VI) (panels (i) and (j)). The different lines with different colors denote different cases (see the legends in panels (e) and (f)). The vertical dotted lines denot the onset, mature, and dissipiation phases.

- Our numerical model allows us to track the accumulation of chemical compounds such as S(VI), NH_4^+, NO_3^- inside of the drops. Additionally, it should be noted that the size dependence of hygroscopicity has no effect on the liquid chemistry occurring in the drops.

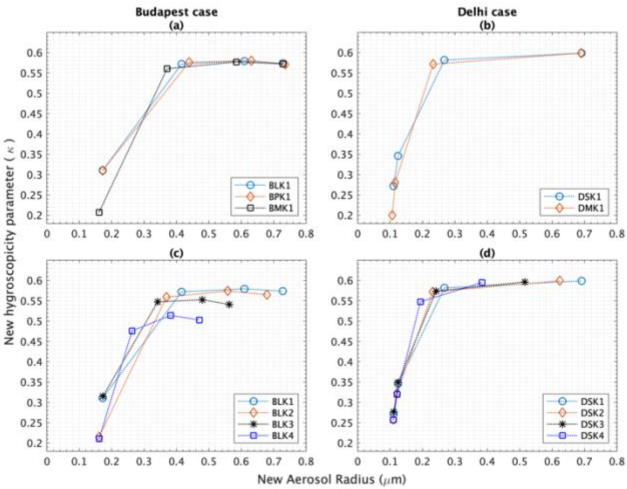

Figure 5: New hygroscopicity parameter for the regenerated aerosol particles at end of fog for the Budapest and Delhi cases.

- The results reveal that liquid chemistry contributes to the broadening of the aerosol size distribution (regenerated aerosol or aging of the aerosol), and significantly increases the hygroscopicity of the regenerated aerosol particles formed after the evaporation of the liquid phase (Figure 5). This increased hygroscopicity may impact the duration of fog dissipation by enhancing the solution effect and helps to promote successive fog event in favourable environmental conditions.

4.3 WRF model results

Thermodynamics and dynamics occur in PBL play a fundamental role in fog formation. In this research, a sensitivity test was accomplished to study how the parameterization of PBL processes impact the accuracy of the fog forecast.

The results of the PBL sensitivity experiment are as follows:

- The parameterization of the PBL schemes significantly impacts the fog microphysics. The amount and the spatial distribution of LWC (liquid water content) are very sensitive on the applied PBL schemes. While QNSE scheme results in unrealistic early formation of the fog (and too large LWC), the duration of the fog is rather short if the non-local schemes (YSU and ACM2) are applied. Although even the MYNN3 scheme results in too early dissipation of the fog, the results suggests that MYNN3 is well suitable for fog prediction over Hungary.
- Unfortunately, all the simulation results (independently of the applied PBL scheme) show the dissipation of the fog starts too early (Pithani et al., 2019, 2018). The reason of this is not known. Because not only PBL processes, but the interaction between the atmosphere and the soil, and the effect of the radiation can be also decisive further research is required to solve this problem.

Publications list

Published papers related to PhD topic:

- **<u>Jeevan Kumar</u>**, B., Geresdi, I., Ghude, S.D., Salma, I., 2021. Numerical simulation of the microphysics and liquid chemical processes occur in fog using size resolving bin scheme. Atmos. Res. 266, 105972. https://doi.org/10.1016/j.atmosres.2021.105972

- Még nem nagykorú, de már adatbázis: Meteorológiai állomás a botanikus kert szélárnyékában – Az Országos Meteorológiai Intézet Szakmai Tájékoztatója 66. : 1. Pp. 11.-18.. (2021) publication link
- Assessment of WRF planetary boundary layer (PBL) schemes in the simulation of Fog events over Hungary (submitted).

List conference presentations related to PhD topic

- Scavenging of submicron size particles by drops in fog - Pune, India. International Conference on Clouds and Precipitation 2021
- Numerical Simulation of Microphysics and Liquid Chemistry in Fog: A Moving Bin Scheme Approach - United States of America AMS 101st Annual Meeting: 13th Symposium on Aerosol - Cloud - Climate Interactions, 23rd Conference on Atmospheric Chemistry
- Scavenging of sub-micron particles by drops in fog Visegrad, Hungary XIV. Magyar Aeroszol konferencia presentation link

- Fog and Smog experience over Northern India in Wintertime. Cluj Napoca, Romania International Conference "Air, and Water - Components of the Environment