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A novel approach for the characterisation of transport and optical properties of aerosol particles near sources – Part II: Microphysics–chemistry-transport model development and application

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ABSTRACT

A new high-resolution microphysics–chemistry-transport model (LES-AOP) was developed and applied for the investigation of aerosol transformation and transport in the vicinity of a livestock facility in northern Germany (PLUS1 field campaign). The model is an extension of a Large-Eddy Simulation (LES) model. The PLUS1 field campaign included the first deployment of the new eye-safe scanning aerosol lidar system of the University of Hohenheim. In a combined approach, model and lidar results were used to characterise a faint aerosol source. The farm plume structure was investigated and the absolute value of its particle backscatter coefficient was determined. Aerosol optical properties were predicted on spatial and temporal resolutions below 100 m and 1 min, upon initialisation by measured meteorological and size-resolved particulate matter mass concentration and composition data. Faint aerosol plumes corresponding to a particle backscatter coefficient down to 10^{-6} sr^{-1} m^{-1} were measured and realistically simulated. Budget-related quantities such as the emission flux and change of the particulate matter mass, were estimated from model results and ground measurements.

1. Introduction

Atmospheric aerosols are part of biogeochemical cycles, influence the hydrological cycle and climate, provide a medium for atmospheric chemistry and contribute to air pollution (Pöschl, 2005). Through aerosol formation, evolvement and removal these impacts are influenced by aerosol microphysical phenomena, atmospheric dynamics and surface processes (e.g. Raes et al., 2000). Aerosol properties are strongly influenced by number and size distributions, which can be represented by a number of log-normally distributed modes, which in turn are formed in processes each determined by characteristic times (Raes et al., 2000; Pöschl, 2005). The main goal of aerosol modelling is to establish a detailed description of the aerosol number and size distributions and composition. Aerosol particle number size distribution (NSD) and composition undergo rapid changes close to particle sources, i.e. up to a few kilometers and minutes from emission. Processes close to sources influence aerosol properties on large scales, but cannot be resolved in large spatial scale models (e.g. Ackermann et al., 1998; Stier et al., 2005) as being sub-grid scale. These have to be addressed by small-scale models, e.g. urban scale and below, and eventually, in parameterised form in large-scale models. Fugitive dust is emitted from cropland, which becomes significant when soils are bare and dry (e.g. Lammel et al., 2003; Chen et al., 2007). Dust has been identified as one of the main uncertainties associated with aerosol sources and properties (Penner et al., 2001). The mineral dust represents generally the main single component of the aerosol's coarse fraction in Europe (Putaud et al., 2004). Investigations into emissions from livestock production have so far mostly concentrated on gases, in particular ammonia, which is an aerosol precursor gas (e.g. Aneja et al., 2001; Baek et al., 2004), and more recently on toxicological hazards, such as endotoxins (e.g. Müller and Wieser, 1987; Seedorf et al., 1998). Primary emissions of particulate matter, however, have hardly been studied. Very few characterisations of aerosol emissions from livestock farming have been reported (Lammel et al., 2004; Martin et al., 2008). Emission factors for primary particulate matter from livestock farming used

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for inventories (e.g. IIASA, 2001) are based on indoor measurements and ventilation rates (Takai et al., 1998; Seedorf and Hartung, 2001; Haeussermann et al., 2008) and considered to be very uncertain. To better understand these emissions, and the changes on composition and particle size distribution that aerosol undergo near their source, studies are needed that involve the analysis of both model and measurement results.

The retrieval of aerosol properties from lidar observations is an ill-posed problem, as different size-resolved aerosol compositions (i.e. different number concentrations by size and component) might generate the same lidar signal. Multi-wavelength lidar systems can provide information about the aerosol size distribution (e.g. Jagodnicka et al., 2009; Veselovskii et al., 2009). Additional information about the aerosol composition might also be inferred from multi-wavelength lidar observations, but this is limited by the number of wavelengths of the system. Depending on the intended application, the size-resolved aerosol composition might be needed in a detail beyond the reach of a particular multi-wavelength lidar system. This was illustrated by Kahnert (2009), who showed that 3-wavelength lidar observations alone did not contain sufficient information to accurately retrieve a size-resolved aerosol composition as described by the model in question. Therefore, model predictions are necessary to better determine the size-resolved aerosol composition, by introducing additional information about the background aerosol and precursor gases.

Previous research about lidar measurements of agricultural aerosol sources is scarce. In order to characterise agricultural emissions, the high spatial and temporal resolution of the lidar offer capabilities that arrays of PM samplers, nevertheless how complex, cannot match (Holmén et al., 1998, 2001a,b). However, lidar measurements are only indirect measurements of the aerosol NSD and composition. Eye-safe lidar systems able to retrieve such quantities have become available in recent years (e.g. Mayor and Spuler, 2004; Pal et al., in press). Faint emissions pose a big challenge for lidar retrievals, but the relative signal intensity from aerosol particles from livestock farm could be detected close to the source (Hartung et al., 1998). Another challenge for lidar retrievals is to provide aerosol optical properties in absolute units. However, for a variety of systems quantitative results concerning aerosol properties have been achieved using sophisticated inversion techniques (e.g. Wulfmeyer and Feingold, 2000). The algorithm for the new mobile eye-safe 3D-scanning aerosol lidar of the Institute for Physics and Meteorology (IPM) of the University of Hohenheim (UHOH) has the capacity to derive absolute values for the particle backscatter coefficient (Behrendt et al., 2005; Pal et al., 2006; Pal, 2009).

The Large-Eddy Simulation (LES) approach has a spatial and temporal resolution comparable with (aerosol backscatter) lidar measurements (Mayor, 2001; Mayor et al., 2003). When combining an aerosol model and LES the high computational demand of the LES is limiting. This leads to either a simple approach to aerosol optical properties (Mayor et al., 2003), or to a comprehensive aerosol module with a reduced spatial representation of the LES model (by reducing the model spatial resolution or the model dimensionality, e.g. Feingold and Kreidenweis, 2002). Forcing a LES model to resemble simple but realistic atmospheric conditions is a challenge by itself (Mayor et al., 2002).

Simulation of LES combined with (aerosol backscatter) lidar observations have previously been performed by Mayor (2001) and Mayor et al. (2003). They compared the wind field structures derived from lidar measurements (described in Mayor and Eloranta, 2001) with an estimate from a LES of a passive tracer. They were interested only “in the patterns of the in homogeneities of relative aerosol scattering created by turbulence” (Mayor et al., 2003) and neither lidar nor LES provided absolute values for the particle backscatter coefficient. In their LES simulations the relative value of the particle backscatter coefficient was estimated as a function of the relative humidity and aerosol particle concentration (simulated as a passive tracer), where the relation between the relative particle backscatter coefficient and the relative humidity being estimated by best fit to experimental data.

This work differs from Mayor (2001) and Mayor et al. (2003) beyond the differences between the respective LES models. In this work, a more detailed description of the aerosol NSD and composition is used, which allows to estimate the absolute value of the particle backscatter coefficient. In addition, the target application of the model demanded a more sophisticated boundary recycling scheme.

The development and first results of a combined approach of LES and lidar (see also Pal et al., in press) to predict and measure the optical properties of aerosol from a faint source, and a field campaign (PLUS1) on the vicinity of a livestock farm in northern Germany, are described in a two part paper. The companion paper (Pal et al., in press, hereafter referred to as part I) describes the characteristics of the new UHOH mobile eye-safe 3D-scanning aerosol lidar, along with the lidar signal calibration technique and lidar measurements during the PLUS1MC. This paper (part II) presents the basic components of a new high-resolution microphysics–chemistry-transport model (LES-AOP), the ground measurements during the PLUS1MC, and the comparison between the simulated and lidar-derived aerosol optical properties. The LES-AOP model and deployment of the instruments during PLUS1 are described on Section 2. Section 3 summarises the experimental results of PLUS1. Section 4 describes the simulated and lidar-derived aerosol optical properties and their comparison. Finally, conclusions are drawn in Section 5.

2. Methods used

Under several assumptions, lidar signal can be inverted to determine the particle backscatter coefficient (Fernald et al., 1972; Klett, 1981, 1985; Fernald, 1984; Pal et al., 2008). The algorithm for the new UHOH scanning aerosol lidar system has the capacity to derive absolute values for the particle backscatter coefficient, including error estimates part I. The particle backscatter coefficient was therefore selected as the parameter for the comparison between model and lidar results.

In order to match the high spatial and temporal resolution of the UHOH scanning aerosol lidar system a new high-resolution chemistry transport model (LES-AOP) was developed, based on a LES model (Chlond, 1992, 1998a,b). The LES-AOP model was designed to simulate particle backscatter coefficient on the temporal resolution of the lidar system. The PLUS1 field campaign was designed to provide a data set for demonstrating the combined model and lidar approach. The general description of the LES-AOP model follows. Details about the aerosol representation and load scenarios for the PLUS1 simulations can be found in Section 4.1.

2.1. High-resolution chemistry transport model

2.1.1. LES extension to aerosol transport

The LES model simulates turbulent flow. It calculates explicitly the large-scale three-dimensional motions, capturing the largest and most energetic structures in the flow while parameterising the effect of the sub-grid-scale eddies (Chlond, 1992, 1998a). The LES model uses Boussinesq equations for the wind components, liquid water potential temperature and total water content (Chlond and Wolkau, 2000).

In Cartesian coordinates, the LES formulation for the conservation equation for a scalar quantity ψ_k is: