Evaluation of WRF-Chem model forecasts of a prolonged Sahara dust episode over the Eastern Alps

Kathrin Baumann-Stanzer\textsuperscript{1}, Marion Greilinger\textsuperscript{1}, Anne Kasper-Giebl\textsuperscript{2}, Claudia Flandorfer\textsuperscript{1}, Alexander Hieden\textsuperscript{1}, Christoph Lotteraner\textsuperscript{1}, Martin Ortner\textsuperscript{3}, Johannes Vergeiner\textsuperscript{4}, Gerhard Schauer\textsuperscript{5}, Martin Piringer\textsuperscript{1}

\textsuperscript{1} Central Institute for Meteorology and Geodynamics (ZAMG), Hohe Warte 38, 1190 Vienna, Austria
\textsuperscript{2} Institute of Chemical Technologies and Analytics, Vienna University of Technology, Getreidemarkt 9/164-AC, 1060 Vienna, Austria
\textsuperscript{3} ZAMG Customer Service Carinthia, Flughafenstraße 60, 9020 Klagenfurt, Austria
\textsuperscript{4} ZAMG Customer Service Tyrol and Vorarlberg, Fürstenweg 180, 6020 Innsbruck, Austria
\textsuperscript{5} ZAMG Customer Salzburg and Upper Austria, Freisaalweg 16, 5020 Salzburg, Austria

Abstract

Sahara dust transport causes a substantial natural background to particle matter concentrations in Central Europe. The contributions by Sahara dust are especially well detectable at Alpine mountain sites where many other sources have minor impact. The ability of a chemical weather forecast model to simulate dust transport correctly is of vital interest as there might be cases of serious health effects for the population which may require countermeasures, as with nuclear or industrial accidents, wildfires, pollen, etc. Here we investigate whether the WRF-Chem set-up, which is run operationally for air quality forecasts in Austria, is able to forecast the transport of the Saharan dust cloud in April 2016 towards Central Europe correctly. WRF-Chem simulations

\textsuperscript{1} Fax: 0043 1 36026 74; Tel: 0043 1 36026 2402; email: k.baumann-stanzer@zamg.ac.at
with and without desert dust emissions reveal that 60 to 70% of the dust arriving at the Eastern Alps originate from desert dust emissions whenever PM concentrations are high during the three periods of this event. The measurements and model results deliver a detailed picture of the course of this extraordinary dust event with successive peaks over the Eastern Alpine region. By the presented example of the analysis of a long-lasting Sahara dust event, a structured approach is proposed to step-wise investigate dust peak episodes based on data analysis of representative background sites, source area analysis by means of Lagrangian dispersion modelling as well as based on coupled meteorological and chemistry modelling.

**Keywords:** Saharan dust, Long-range Transport, WRF-Chem model, FLEXPART model, Source-receptor-sensitivity, Ceilometer

**INTRODUCTION**

Almost $2 \times 10^9$ tons of dust are emitted every year from arid and semi-arid areas (Monks et al., 2009). The largest dust source region in the world is North Africa. Ginoux et al. (2012) estimate, based on MODIS dust optical depth images combined with land use information, that North Africa accounts for 55% of global dust emissions with only 8% of anthropogenic origin. The main potential source areas of African dust are Western Sahara and Morocco, Algeria and Tunisia (Salvador et al., 2014). Most of the Saharan dust, about 60%, is transported westward
across the Atlantic and to the Caribbean (Prospero and Mayol-Bracero, 2013), but about 10% of
the desert dust which is entrained into the free atmosphere is transported towards the
Mediterranean Sea and Europe depending on the prevailing large-scale pressure fields and flow
patterns (Schepanski et al., 2016). According to Pey et al. (2013), African dust outbreaks are
more frequent in southern sites across the Mediterranean, from 30 to 37% of the annual days,
whereas they occur less than 20% of the time in Central Europe. Nevertheless, the identification
and quantification of dust transport from desert areas to Europe is relevant as these events can be
classified as “natural events” in the official reporting of air quality measurements to the European
Union. For this purpose, Alpine Observatories render especially valuable data on these dust
clouds as these background measurements are less influenced by other sources as traffic, industry
and domestic burning (e.g. De Angelis and Gaudichet, 1991, Coen et al., 2004).

Besides the analysis of particle matter measurements at mountain tops in the Eastern Alps, the
source regions of the dust clouds and the pathways to the Alpine ridge are of interest to scientists
as well as to authorities planning counter-measures. Sodemann et al. (2006) conclude from ice
core data and trajectory analyses that different transport pathways rather than different source
areas lead to major differences in the chemical signature of the deposited dust. The transport
patterns of dust from the Saharan desert to the Alps can vary substantially from case to case or
even within phases of a single event lasting for several days. In many dust transport studies, back-
trajectories (Ansmann et al., 2003; Schauer et al., 2016; Querol et al., 2009) or ensembles of trajectories (Sodemann et al., 2006; Salvador et al., 2014) are used. Birmili et al. (2010) apply the Lagrangian model FLEXPART in backward mode to investigate transatlantic aerosol transport to the Alps. Column-integrated source-receptor sensitivity (SRS) fields are used to reveal the overall pathway of the air. The SRS fields in the lowest 100 m AGL are interpreted by Birmili et al. (2010) in terms of a “footprint” relationship indicating the dominant source regions. In this study, the same backward modelling approach is used to clarify, whether the dominant source regions are situated in the Saharan desert during the episode under investigation.

Flaounas et al. (2016), Rizza et al. (2017) and Nabavi et al. (2017) present improved source function schemes to estimate dust emissions from arid areas for the on-line coupled chemical and meteorological forecast model WRF-Chem.

Remote sensing data from satellites (e.g. Alpert and Ganor, 2001; Ginoux et al., 2012; Monks et al., 2009; Todd and Cavazos-Guerra, 2016) and lidar profiles (Ansmann et al., 2003; Salvador et al., 2011) are used to monitor the release and transport of dust plumes. In this study, ceilometers are used to help investigate whether a dust plume is primarily advected or is mixed upwards from ground-level sources to the mountain tops by vertical mixing. Ceilometers are aerosol lidars which are operated since decades at airports to detect the cloud base height. The software of modern ceilometers is able to analyse the first few kilometres above ground for the
existence of so-called aerosol layer heights which mark transitions between layers of different aerosol content (Lotteraner and Piringer, 2016).

Based on the measurements at three mountain top sites in the Eastern Alps, combined with aerosol profiles observed with ceilometers and the source region analysis, the evolution of a Sahara dust event in April 2016 is elaborated and the capability of WRF-Chem to forecast this event is demonstrated.

The Saharan dust cloud reached Central Europe on March 31, 2016 and remained there for about 10 days. Enhanced levels of PM10 concentrations were most obvious on mountain stations as well as at many stations of the routine air quality monitoring network across Austria. The proof that the enhanced PM10 concentrations originated from the Sahara is carried out based on observations and models. The novel aspect of this research is the step-wise analysis of in-situ as well as remote sensing measurements and the application of forward and back-ward modelling in order to get a deeper understanding of the evolution of a dust event.

All in all, this study reveals how measurements and analysis tools can be used to investigate exceptional air pollution events caused by long-range transport.

METHODS

Site Description
Particulate matter (PM) measurements at three Austrian mountain stations Feuerkogel, Sonnblick and Dobratsch, situated in a north to south transect across the Alps, are used to evaluate the WRF-Chem model performance. The global GAW station Sonnblick is located at the main alpine ridge, while Feuerkogel and Dobratsch are situated further north and south, respectively. Three ceilometers located in Salzburg, Radstadt and Nötsch, were used to measure the aerosol structure and to better interpret observed PM concentrations. Fig. 1 displays the location of the sites.

The continuously manned Sonnblick Observatory (SBO) is one of 31 global stations of the Global Atmospheric Watch Program (GAW), situated at the summit of Mt. Sonnblick at 3,106 m asl in the Austrian Alps (12°57’E, 47°03’N), within the core zone of Hohe Tauern National Park. The peak of Mt. Sonnblick is surrounded by large glacier fields and located approx. 1000 m above the tree line.

The continuously manned meteorological station „Villacher Alpe“ is operated by ZAMG on top of Mt. Dobratsch (13°40’E, 46°36’N) in 2,117 m asl since 1994. The station was additionally equipped with particle matter measurements in October 2015.

The continuously manned meteorological station on top of Feuerkogel (13°43’E, 47°49’N) at 1,618 m asl was put into operation in 1992 and is additionally equipped with particle matter measurements since January 2016.
Particulate matter measurements

The size fraction PM10 (particulate matter <=10 µm aerodynamic diameter) is determined with a Grimm EDM 180 Dust Monitor at the top of Feuerkogel and with a SHARP-Monitor (Thermo Scientific, Sharp 5030) at the top of Dobratsch.

At the Sonnblick Observatory, particulate matter is sampled with a SHARP-Monitor (Thermo Scientific, Sharp 5030) via a heated whole air inlet, which is recommended for high alpine mountain sites according to the GAW guidelines (WMO/GAW Report No. 153, 2003). The upper cut-off size of the inlet is 20 µm at a wind speed of 20 ms⁻¹, but for particles > 1 µm, losses will occur (Schauer et al. 2016). Thus, concentrations determined during the transport of mineral dust have to be regarded as a lower limit to actual conditions at the site. On the other hand no local sources of PM with particle diameters larger than 10 µm can be expected at the site as the soil is rock or ice/snow covered. Transport of PM with particle diameters larger than 10 µm to the top of Sonnblick from sources at the valley bottom is unlikely as these particles are removed at lower levels due to their deposition velocity. Thus the PM mass concentrations at Sonnblick are compared with PM10 concentrations from model output or other sites. Further, PM measurements at Sonnblick comprise the total number concentration using a condensation particle counter (TSI CPC 3022A), and particle size distribution is measured in the size range between 0.3 and >2.5 µm via an optical particle counter (Klotz, TCC-3). Optical aerosol properties, especially the single scattering albedo (SSA) and the absorption coefficient (ABS), are
measured using a three-wavelengths polar Nephelometer (Ecotech Aurora 4000) and a seven-wavelengths Aethalometer (Magee Scientific AE 33). All measurements are reported as 30 min averages and aerosol concentrations given are calculated for 273 K and 1013 mbar. The wavelength dependence of the SSA and the ABS are investigated. The wavelength $\lambda$ dependence of both parameters is fitted with a power-law dependence to obtain the respective exponents, $\alpha_{SSA}$ and $\alpha_{ABS}$. The equation is shown exemplarily for the exponent $\alpha_{SSA}$:

$$SSA = b_{SSA} \times \lambda^{-\alpha_{SSA}}$$

The wavelength dependence of the SSA is used to identify long range transport of mineral dust, e.g. originating from the Sahara (Coen et al., 2003).

In order to detect dependencies of pollutant distributions on flow patterns, daily mean PM10 concentrations at Sonnblick, Dobratsch and Feuerkogel are analysed according to the large-scale flow between January 14, 2016 and December 31, 2016 based on the operational synoptic weather circulation types classification implemented at the national weather service Austria ZAMG (Classification of Atmospheric Circulation Patterns representative for the Alpine area model output data) in Fig. 2. Daily mean PM concentrations are calculated by averaging time-series of half hourly concentration values. A minimum of 75% of data availability per day is required to render a valid daily average.
Ceilometer measurements

The ceilometer CL51 employs a diode laser lidar technology. Laser pulses with a wavelength of 910 ± 10 nm are sent out in a vertical or near-vertical direction. The measurement range of the instrument extends from approximately 50 m above the ceilometer site, where a sufficient overlap of emitted and backscattered laser signals is given (Wagner and Schäfer 2015), up to 15 km above ground.

The base heights of up to three cloud layers as well as up to three aerosol-layer heights are detected from the observed backscatter profiles by applying the so-called gradient method described in Emeis et al. (2007). An improved algorithm to determine the mixing height from ceilometer aerosol layer heights is described in Lotteraner and Piringer (2016).

Source Region Analysis

The Lagrangian particle model FLEXPART (Stohl et al., 2002), a stochastic particle dispersion model with turbulence and convection parameterisations is used to identify the source regions of the air arriving at the mountain top sites. Lagrangian particle models usually release a high number of particles at defined source locations, compute their forward trajectories according to the three-dimensional wind fields with regard to all linear processes influencing the mass attached to these particles, i.e. advection, diffusion, convective mixing, dry and wet deposition. Non-linear chemical reactions cannot be simulated with this modelling approach. By reversing the sign of the advection, the trajectories can easily be calculated backward, starting in this case
from the receptor points e.g. measuring stations. This methodology, described in detail by Seibert and Frank (2004), is used in the presented study, applying FLEXPART in backward mode in order to detect the source regions for the air which is probed by the instruments at the mountain sites during a particular measuring period (of typically 3 hours length). The model results are presented as spatial distributions of the so-called source-receptor-sensitivities (SRS). The values of this model parameter can be interpreted as spatial information indicating those areas where a larger or smaller proportion of the air at the respective site came from.

In this study, FLEXPART is applied in backward mode based on model-level analyses from the European Centre for Medium Range Weather Forecast with 1°x1° resolution and 60 vertical layers. In contrast to Birmili et al. (2010), source-receptor sensitivity fields are evaluated for the lowest 2 km AGL. This setting is optimized for the application of Saharan dust transport detection. The layer between 0 m and 2000 m AGL is considered in order to capture contributions from sources near ground, dust uptake within the major part of the atmospheric boundary layer during day-time as well as elevated dust layers within the residual layer above the nocturnal boundary layer. Test runs revealed that SRS fields are similar for 0 m to 1500 m AGL or for 0 m to 2500 m AGL, but significantly change when near-ground levels are considered only as in Birmili et al. (2010). The results of these backward model runs in Fig. 9 indicate the origin of air
masses which arrive at the mountain stations north, south and at the Alpine ridge within a 3-
hourly time interval.

Chemical Weather Forecast

WRF-Chem (Grell et al., 2005) is the Weather Research and Forecasting (WRF) model coupled
with chemistry. This on-line coupled model simulates the emission, transport, mixing, and
chemical transformation of trace gases and aerosols simultaneously with the meteorology. In the
model set-up for this study, the direct (radiation), indirect (cloud, precipitation) and semi-direct
(wind, temperature, humidity) feedbacks/effects between meteorology and air chemistry are
included. WRF-Chem version 3.4.1 is applied with the PBL scheme developed by Yonsei
University, the cloud microphysics scheme described by Morrison and Milbrandt (2015), NOAH
land surface model (Niu et al., 2011) and the RRTMG scheme for long wave and short wave
radiation (Iacono, 2008). The RADM2 chemical mechanism (Stockwell et al., 1990) is used for
gas phase chemistry. Biogenic emissions are considered by the MEGAN approach developed by
Guenther et al. (2006). The latest anthropogenic emission inventories provided by the Austrian
federal governments are combined with emission data provided by TNO (Visschedijk et al., 2007)
and EMEP (http://www.ceip.at/ceip).

Sahara dust is simulated in this study using the aerosol module including aqueous reactions
MADE/SORGRAM (Schell et al., 2001) with dust emission option. In the WRF-Chem model
output, the parameter \( DUST_{\text{col}} \) is the sum of the vertical integral (up to 50 hPa) of the coarse soil derived aerosols and the coarse soil derived aerosols in clouds.

In the current study, the meteorological fields are based on ECMWF forecasts with 0.125°x0.125° horizontal resolution and 138 vertical layers. The episode has been simulated with WRF-Chem runs for 24 hours including a daily initialization of the meteorological fields. The horizontal resolution is 12 km for the area of Europe and North Africa (see model domain in Fig. 10). PM10 concentrations simulated with WRF-Chem are compared to observations, and model runs with and without modelled desert dust emissions are used to investigate the impact of Saharan dust transport on the PM10 concentrations in the Alpine region.

RESULTS AND DISCUSSION

This section starts with an overall analysis of the particle matter concentrations found at the mountain top stations in different flow regimes. It continues describing the evolution of PM mass concentrations during the Sahara dust episode and compares the measurements with observed aerosol profiles from nearby ceilometers. This is followed by a source region analysis based on FLEXPART backward runs. This observed data analysis is then contrasted with the forecast of the Sahara dust event based on WRF-Chem concentration fields. Finally, the forecast is evaluated comparing it with the aerosol measurements. In this way, a composite picture of this extraordinary Sahara dust event is achieved.
Particle matter at mountain tops in different flow regimes

In the following, the occurrence of events with increased PM concentrations at the three mountain top stations is investigated according to the prevailing synoptic conditions. Usually, pollution levels at mountain tops are quite low. Pollutants conveyed by long-range transport at higher levels or events with pollutants mixed from the valley floors to the mountain tops typically under convective conditions causing enhanced concentrations are therefore often clearly visible.

Analyses of predominant flow patterns for the year 2016 as well as on a long term average from 2000 to 2016 reveal a predominance of south-westerly flows and weather patterns with weak winds. Hence, the statistical analysis of daily mean PM10 concentrations at mountain stations is most significant for these two large-scale flow patterns (Fig. 2). During south-westerly to south-easterly flows, PM10 concentrations on average tend to be slightly higher at Dobratsch, on the south side of the Alpine main crest than at Feuerkogel, on the north side of the Alps. Besides of the impact of local sources (wood burning in villages), polluted air from industrial areas in the Po Valley is transported to the southern Alpine crest under southerly flow conditions while the northern Alpine crest is less affected in these cases.

On the other hand, PM10 concentrations tend to be higher at Feuerkogel north of the Alps than at Dobratsch south of the Alps during northerly flows as is seen from the upper whiskers and outliers in the columns of north-westerly, northerly and north-easterly flows (Fig. 2). In these cases, the northern Alpine crest is exposed to long-range transport from various industrial areas in
Europe. However, it must be mentioned that except for south-westerly flows, sample sizes for weather patterns with weak winds and north-westerly flows are too small to achieve statistical representativeness. Thus, these dependencies of PM10 concentrations and distributions on flow patterns can only be evaluated qualitatively.

Four days with high daily PM10 concentrations at all three mountain sites are highlighted in color in Fig. 2. The on-line monitoring of mineral dust, which is performed at the Sonnblick, indicates the occurrence of dust events during those days. This monitoring follows the approach of Coen et al. (2004) and evaluates the wavelength dependence of the single scattering albedo (SSA) in the range from 450 to 635 nm. The SSA is calculated via the scattering and absorption coefficients determined at the site. A detailed description of the index which is used to identify Saharan dust events from these measurements can be found in Schauer et al. (2016). The following analyses focus on the episode leading to the maximum daily mean PM10 concentrations observed at the Austrian mountain stations on April 5th, 2016.

**Evolution of PM mass concentrations during the episode**

In the following, the evolution of the PM concentrations at the three mountain tops during the period of investigation is described as revealed by the measurements. Fig. 3 depicts the temporal evolution of PM10 concentrations at Dobratsch, Feuerkogel and PM concentrations at Sonnblick for the Saharan dust episode from March 31, 2016 to April 7, 2016.
The temporal evolution of the PM concentrations at all three stations allows a rough separation of the event in three episodes. Episode 1 spanned from March 31 12:00 UTC to April 3 12:00 UTC, occurring almost simultaneously at the three stations, merely with different intensities. Episode 2 spanned from April 3 12:00 UTC to approximately April 5 00:00 UTC with a delay of the increase of PM mass at Dobratsch of about 12 hours compared to Sonnblick and Feuerkogel. Episode 3 started almost simultaneously at all three stations on April 5 00:00 UTC and lasts until April 6 00:00 UTC at Feuerkogel, but a few days longer, approximately until April 7 12:00 UTC, at Sonnblick and Dobratsch.

During all episodes, the highest PM concentrations are observed at Sonnblick. Saharan dust is transported at high elevations and over large distances via synoptic wind patterns. Thus, related PM concentrations are expected to be highest at high elevations, such as at Sonnblick (3.106 m asl). Due to vertical mixing, the long-range transported PM is mixed down, causing lower concentrations at lower elevations like at Feuerkogel (1.602 m asl) and Dobratsch (2.160 m asl).

The episodic evolution of PM mass can be explained via the prevailing synoptic wind pattern during that time. For episode 1, a low pressure system over the Baleares transported the Saharan dust loaded air from the Western Sahara towards Europe. This low pressure system moved towards the Mediterranean, thereby weakening in intensity. Still Saharan dust was blown up into the air, but this time from the Eastern Sahara. Austria was under the influence of high pressure
during that time, carrying the eroded dust towards Austria during episode 2. Simultaneously a new low pressure system was formed over the Iberian Peninsula moving south towards Algeria, thereby intensifying. This low pressure system again mixed Western Saharan dust into the air which was then transported towards Austria during episode 3.

The meteorological analysis of the investigated episode reveals different Saharan dust source regions for the episodes. Various aerosol properties measured at Sonnblick were used to investigate potential differences in the aerosol composition, although no chemical analysis was performed for the different episodes.

First, the wavelength dependence of the SSA is used to identify long range transport of mineral dust. Calculating the SSA exponent $\alpha_{\text{SSA}}$, the change in sign of $\alpha_{\text{SSA}}$ to negative values is used as an indication for mineral dust affecting the site. Regarding the whole episode, a median $\alpha_{\text{SSA}}$ of -0.17 was obtained. This indicates that the majority of PM mass is represented by Saharan dust (Schauer et al., 2016). Median values of $\alpha_{\text{SSA}}$ for the three sub-episodes are near to -0.19 (compare Fig. 4 a). The wavelength dependence of the absorption coefficient is expressed by calculating the absorption exponent $\alpha_{\text{ABS}}$. For the whole episode the median $\alpha_{\text{ABS}}$ is 1.66, while median values of the three sub-episodes are 1.54, 1.80 and 1.88, respectively (compare Fig. 4 b). Fialho et al. (2006) reported an $\alpha_{\text{ABS}}$ of 4.0 as typical for pure dust, whereas Schauer et al. (2016) used $\alpha_{\text{ABS}}$ of 3.0 for Saharan dust and $\alpha_{\text{ABS}}$ of 1.3 for
wildfire. For the Saharan dust episode investigated here $\alpha_{\text{ABS}}$ values for Saharan dust were much lower and, regarding the observed range of $\alpha_{\text{ABS}}$ values in literature, results for the sub-episodes are not markedly different. Minor differences can be related to differences in the source region, but also to the amount of Saharan dust within the aerosol influencing the coloring and hence the absorption properties of the particles.

Particle size distributions are used in the following to investigate potential differences in the episodes of the investigated Saharan dust event.

Secondly, particle size distributions are used here to investigate potential differences in the different sub-episodes of the investigated Saharan dust event. The temporal evolution of the particle numbers of particles greater than 2.5 µm (coarse particles) and the total particle number (denoted as CP-count) is displayed in Fig. 5 together with PM mass. PM mass is highly correlated to the number concentrations of coarse particles ($R^2 = 0.997$). In contrast to this, the CP-count showed little variation over all three sub-episodes and no correlation with either PM mass or the number concentration of coarse particles ($R^2 = 0.02$ and $R^2 = 0.01$, respectively).

If the particle number of the coarse fraction (optical diameter $>2.5$ µm) is compared to the number of particles with an optical diameter $>0.3$µm (compare Fig. 6), the contribution of coarse particles to number concentrations of particles $>0.3$µm is almost similar for Episode 1 and 2 with mean values of 5.0% and 5.2%, respectively, whereas for Episode 3 the ratio increased to a value
of 6.7%. Schauer et al. (2016) found values between 1.6-4.3% (representing the 10th and 90th percentile) for a Saharan dust episode in August 2013.

**Observed aerosol profiles**

Besides of the in-situ measurements at the three mountain stations, the available remote-sensing data from ceilometers give further insight in the vertical distribution of aerosol density. The observations during the period of interest are presented in the following. The ceilometer at Nötsch is situated at the base of Dobratsch. Radstadt is situated in the valley directly north of the main Alpine chain. Salzburg is situated at the northern edge of the Eastern Alps comparable to the northern side of Feuerkogel.

The arrival of two dust clouds can be discerned from the backscatter plots of the three ceilometers in Fig. 7 and Fig. 8. The first starts in the evening hours of March 31, the second just before midnight on April 3. The color coding in the figures is a measure of aerosol backscatter intensity, increasing from blue to full green. Dark blue areas indicate no data. Such areas are found most often above a cloud layer, shown by yellow and red lines near an aerosol layer height (black dotted lines). Red areas indicate rain. The first dust cloud on March 31 arrives almost at the same time at all three sites (Fig. 7). The enhancement of aerosols starts above 2 km AGL and propagates quite rapidly downwards. North of the main Alpine chain, clouds prevent the detection of the dust cloud on April 1 first at Salzburg (Fig. 7 a), later at Radstadt (Fig. 7 b).
At Nötsch south of the Alpine chain, clouds are present at forenoon on April 2 (Fig. 7 c). Especially from the plots at Radstadt, the impression of a persistent dust cloud above 3 km on April 2 and April 3 evolves (Fig. 7 b). In the late evening on April 3, the second much stronger dust cloud arrives at ground level, first at Salzburg (Fig. 8 a), later at Radstadt (Fig. 8 b) and Nötsch (Fig. 8 c). This is in good accordance with the contemporaneous increase of PM observed at the mountain stations at the beginning of episode 2 (Fig. 3). In this episode, the dust cloud takes much more time to arrive at the surface, especially at Nötsch, where full mixing is observed on April 4 from about noon onwards only (grey line in Fig. 3). April 5 is the peak day of episode 3 as well as of the whole event with uniform spread of dust throughout the boundary layer at all sites with a top around 3 km (Fig. 8). The visibility of the dust cloud ends first in Salzburg and later in Radstadt (Fig. 8 a and b) due to a new cloud layer, with rain in the early morning on April 6. At the same time, the rapid decrease of PM concentrations is observed at Feuerkogel (dashed line in Fig. 3).

At the ceilometer sites Salzburg and Radstadt, cloud layers remain throughout the rest of the investigation period, preventing the detection of the Saharan dust cloud. At Nötsch, the dust layer is still visible throughout April 6 (Fig. 8 c) and April 7 (not shown). A slight decay of aerosol backscatter intensity is visible in the ceilometer profiles within the mixed layer from the afternoon of April 6 onwards (Fig. 8 c) indicating a gradual decrease of aerosol concentrations.
The later finding is in good agreement with the gradual decrease of PM measurements at Sonnblick and Dobratsch in the second half of episode 3 (black and grey lines in Fig. 3).

**Source region analysis**

The origin of the air masses which arrived at the Eastern Alps in the three phases of this event is investigated subsequently. The aim of this part of the investigation is to clarify whether dispersion modeling based on the large scale flow supports the hypothesis that the observed particle matter at the mountain sites during the respective period can be attributed to sources in Northern Africa. For this purpose, the areas where air masses arrive at Sonnblick during selected three hour intervals are identified by applying FLEXPART in backward mode. Fig. 9 (a) depicts the integral of hourly fields of SRS for the layer between 0 m AGL and 2000 m AGL from the five previous days calculated for the time period April 1, 2016 9 UTC to 12 UTC when peak PM10 concentrations of episode 1 were observed at Sonnblick. The numbers in the colored boxes (Fig. 9) give the proportion of air prevailing in the respective area within the previous five days. It has to be considered that air masses passing the blue box around Sonnblick in Fig. 9 partly originate from long-range transport. According to this model result, 22 % of the air masses arriving at Sonnblick originated from the High Atlas and the Algerian Sahara (orange box in Fig. 9 a). A contribution of 7 % of the air masses originates in the eastern Sahara (green box in Fig. 9 a).
The backwards transport modeling for the second period indicates a different result. On April 4, 20% of the air masses can be retraced to the eastern Sahara (green box in Fig. 9 b), whereas only 2% still originate from the western Sahara (orange box in Fig. 9 b).

For the third period, the FLEXPART model result in Fig. 9 c renders that 24% of the air masses originate from the High Atlas and the Algerian Sahara (orange box) and 8% from the eastern Sahara (green box). Thus, the source regions of desert dust varied significantly throughout this event although no clear difference could be detected in the chemical analysis of the aerosol measurements.

**Forecast of an extraordinary Sahara dust event**

Measurements at mountain tops render valuable background concentration data for model evaluation. This is especially the case for long-range transport events as increased air pollution values preferably occur at higher level under these conditions. Therefore, the results of the operational WRF-Chem model runs which are conducted to forecast air quality in Austria are presented here and compared to the measurements in the following section. The ability to forecast long-range transport phenomena is of vital interest as there might be cases of serious health effects for the population which may require countermeasures, as with nuclear or industrial accidents, wildfires, pollen, etc. Here we investigate whether WRF-Chem is able to forecast the transport of the Saharan dust cloud from April 2016 towards Central Europe correctly.
According to the WRF-Chem model results, a dust cloud covered a large part of Central Europe already on April 1, 2016, propagating more to the East on April 2, 2016. The spatial distribution of dust concentration in the whole air column up to the top of the model domain (50 hPa) on April 3, 2016 12 UTC and April 5, 2016 12 UTC calculated with WRF-Chem is shown in Fig. 10 (a) and (b), respectively. According to the model results, the largest concentrations remained over the Algerian Sahara desert until April 4, 2016, most likely indicating sand storm events in this area (see Fig. 10 a). On April 5, 2016, the dust cloud is well seen in the WRF-Chem model output (Fig. 10 b), the area with highest concentrations extending from the western Mediterranean Sea to the eastern Alpine area and covering major parts of eastern Europe to the Black Sea. The dust cloud remains clearly visible in the Alpine area in the WRF-Chem model results until April 8, 2016.

Fig. 10 (c) and Fig. 10 (d) reveal the impact of desert dust emissions on the dust concentrations as forecasted by the model on April 3, 2016 12 UTC and on April 5, 2016 12 UTC, respectively. In the High Atlas region as well as at the Arabian Peninsula, red areas indicate regions were sand storms are taking place. The red shaded region covering the western Mediterranean Sea and Central Europe on the other hand is clearly indicating the position of the Saharan dust cloud which has been transported here during the previous days.

Forecast evaluation
In the following, the operational WRF-Chem model forecasts are evaluated in comparison to the aerosol measurements described in the previous sections. Medium- to long-range transport models in general may not resolve all the details of air pollution as revealed by in-situ observations at stations mainly because the latter may reflect the impact of local sources which are not revealed by the model emission data base.

According to the model results, up to 70% of the particle matter at the mountain tops can be traced back to dust emissions during the first six days of the event (dashed line in Fig. 11). The first PM peak at Sonnblick on April 1, 2016 is well forecasted by the model regarding intensity as well as on-set and temporal evolution (Fig. 11 a). The model forecasts for Dobratsch (Fig. 11 b) and Feuerkogel (Fig. 11 c) also are in good agreement with the observed arrival of the dust cloud during the first episode of the event. The largest discrepancies between the modeled and measured PM concentrations at all three sites are found in the results for April 3 and 4. In contrast to the measurements, the WRF-Chem results indicate a second increase of dust at Sonnblick and Feuerkogel in the beginning of April 3, 2016. The ceilometer profiles at Radstadt clearly reveal that an aerosol layer was present above 3 km above ground at the respective time (compare Fig. 11 a, Fig. 11 c and Fig. 7 b). Obviously this dust layer remains above the mountain tops and therefore has no impact on the in-situ aerosol measurements in the first half of April 3rd. The aerosol profiles observed by the ceilometers at Salzburg and Radstadt (Fig. 8)
reveal a gradually descending aerosol layer during the afternoon of this day and the following night. At Dobratsch, a weak increase is visible in the model results on the same day around noon. On the other hand, an intense increase of PM is recorded at all three mountain stations at the beginning of April 4, 2018 (Fig. 11), while the concentration values forecasted with WRF-Chem remain low until the night to April, 5th.

No straightforward explanation for the differences between modeled and observed PM concentration on April 3 and April 4, 2016 can be given based on aerosol characterization. The simulated meteorological conditions on both days, dominated by a high pressure system over the Alpine area with large scale subsidence of dry upper level air, are in close agreement to the in-situ and the remote sensing observations. The only significant difference is that the model simulation indicates a small amount of precipitation at Sonnblick which is not confirmed by the observations. As this gives no explanation for the differences between modelled and measured PM concentrations on April 4, it is concluded that either the dust emissions due to wind erosion or the transport of the dust cloud leading to the observed increase of PM concentrations on April 4, 2016 was underestimated by the model.

A closer analysis of the model results for Dobratsch reveals that the simulated PM concentrations follow a daily cycle with increased values at noon and low values at night during the whole period of investigation (Fig. 11 b). This is a clear indication that the model simulates
boundary layer air enriched with particle matter from emission sources in the area is reaching the site during day-time.

Finally, the time of the third and most intense arrival of dust at Sonnblick and Feuerkogel on April 5, 2016 is forecasted very well by WRF-Chem (Fig. 11 a and c). The model also simulates successfully this dust event at the northern side of the Alpine ridge, at Feuerkogel (Fig. 11 c) in terms of magnitude as well as the sudden decrease of PM concentrations caused by the change of air masses with a frontal passage taking place on April, 6\textsuperscript{th} midnight. Still the PM concentrations are clearly underestimated at Sonnblick (Fig. 11 a) and at Dobratsch (Fig. 11 b). According to the ceilometer measurements the aerosol layer was most pronounced and persisted at the southern side of the Alpine ridge after the end of the investigated period (Fig. 8 c). The percentage of dust due to wind erosion in the model results (dashed line in Fig. 11) decreases gradually at the end of the period. This is in good agreement with the PM concentrations observed at the mountain sites.

**CONCLUSIONS**

The measurements and model results presented in the previous chapters reveal that dust clouds which originate from sand storms in Northern Africa significantly contribute to particle matter concentrations over the Eastern Alps. The large scale flow regime as determined with weather classification from model output gives a relatively good first indication whether peak
concentrations of particle matter at mountain tops in the Eastern Alps might be influenced by long-range transport from North Africa. In the statistical analysis of the PM measurements at three mountain sites for the year 2016 in Fig. 2, the highest daily mean concentrations are observed under south-westerly flow conditions. The maximum PM concentrations of the year at the three mountain tops occurred in the first week of April 2016. This event can be separated into three episodes with waves of dust arriving at the mountain tops as depicted in Fig. 3.

The on-set of Sahara dust import to the Alps in the first episode, the maximum PM peak during the third episode and the end of the Sahara dust event in April 2016 are in general successfully forecasted with the model WRF-Chem. The dust emissions in the Algerian Sahara desert due to stormy winds as well as the transport across the Mediterranean Sea to Central Europe are reproduced in the model considerably well. During the second episode of the event, modeled and observed PM mass concentrations differ significantly in amount as well as in the temporal evolution (Fig. 11). On-line characterization of the aerosols based at Sonnblick Observatory renders no explanation for differences between the three episodes. Uncertainties in the estimation of the dust emission based on the forecasted surface wind speed in the desert area, deviations in the simulated regional flow fields as well as uncertainties in the representation of the atmospheric boundary layer close to the mountain tops in the model seem to be responsible for these differences. The evolution of the aerosol profiles observed by ceilometers during the event
illustrate the complexity of the interacting processes, descending layers of dust from higher levels mixing into the atmospheric boundary layer in the course of the days (Fig. 7 and Fig. 8). The results of the FLEXPART backward model runs render insight in the most dominant source regions of the PM10 concentrations at the mountain tops and even allow the approximate quantification of the proportion of air masses originating from predefined areas. The quantification based on the SRS fields was performed for the first time in the course of the presented study. This methodology will be further elaborated in order to support the general interpretation of air quality measurements.

The good agreement between model results and observations in the first two days of the event as well as the successfully forecasted PM10 concentrations on the last day of the event show that the parameterizations and assumptions used in the operational WRF-Chem runs are appropriate to forecast dust storm emission, transport as well as deposition of particle matter at the mountain sites. Therefore, it is concluded that the estimation of up to 70% of contribution of Sahara dust to the observed PM concentrations during the event in April 2016 as simulated by WRF-Chem (Fig. 11) is plausible.

This case study demonstrates that the combination of the used measurements and analysis tools is a good basis for the interpretation of exceptional air pollution events caused by long-range transport as Sahara dust transport to the Alps. In general, peak episodes observed at background
sites can be investigated by the proposed structured approach involving a detailed analysis of the observational data, a source area analysis by means of Lagrangian dispersion modelling as well as meteorological and chemistry modelling.

In the case of increased PM concentrations, the identification of long-range transport is of particular relevance as occurrences of dust import from arid areas can be classified as “natural events” in the official reporting of air quality measurements to the European Union and do not add to the number of PM10 daily mean threshold exceedances.

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REFERENCES


Figure Captions

**Fig. 1.** Detail map of Eastern Alps in Austria with mountain top air quality stations (triangles) and meteorological stations and ceilometer CL51 sites (circles).

**Fig. 2.** Statistical analysis (boxplot) of daily mean PM10 concentrations at the mountain tops of Sonnblick (Son), Dobratsch (Dob) and Feuerkogel (Feu) between January 14, 2016 and December 31, 2016 separated according to the large-scale flow (Classification of Atmospheric Circulation Patterns representative for the Alpine area deduced from model output). XX represents all days when weak pressure gradients were prevailing and no dominant synoptic-scale flow direction is discernible. The bottom and top of the box depict the lower quartile (Q1) and upper quartile (Q3), the band inside the box is the median (M). The lines extending vertically from the boxes (whiskers) indicate the minimum and maximum values (except for outliers). Outliers (O) are defined as any points larger than Q3 + 1.5*IQR or lower then Q1 - 1.5*IQR, where IQR is the inter quartile range defined as IQR=Q3-Q1. Outlier values larger than 50 μg m⁻³ PM10 are marked in colours and the respective day is given in the legend on the right side of the figure.

**Fig. 3.** Temporal evolution of PM concentrations at Sonnblick, Dobratsch and Feuerkogel from March 31, 2016 0 UTC to April 9, 2016 0 UTC. As explained in the text no defined size cut can be reported for measurements at Sonnblick, whereas PM10 was determined at Dobratsch and Feuerkogel. Vertical grey lines indicate the three different episodes of this event.

**Fig. 4.** Boxplot of α_SSA (left) and α_ABS (right) for the whole Saharan dust episode from March 31, 2016 to April 7, 2016 as well as for the three episodes (E1 from March 31 12:00 UTC to April 3 11:30 UTC, E2 from April 3 12:00 UTC to April 4 23:30 UTC and E3 from April 5 00:00 UTC to April 7 12:00 UTC).

**Fig. 5.** PM mass in μg m⁻³ (black) as well as particle numbers per cm³ of particles > 2.5 μm optical diameter (dark grey) and condensation particle count (CP-count) representing the total particle number (light grey) of the Saharan dust event from March 31, 2016 0 UTC to April 8, 2016 0 UTC. Vertical grey lines indicate the three different episodes of this event.

**Fig. 6.** Particle numbers per cm³ of particles > 2.5 μm optical diameter (blue) and particles > 0.3 μm optical diameter (pink) as well as the ratio of those two (black) for the Saharan dust event from March 31, 2016 0 UTC to April 8, 2016 0 UTC. Vertical grey lines indicate the three different episodes of this event.
Fig. 7. Time-height cross sections of ceilometer aerosol backscatter intensity a) at Salzburg, b) at Radstadt and c) at Nötsch between March 31, 2016 0 UTC and April 3, 2016 12 UTC. The vertical white line indicates the beginning of the first episode of this event.

Fig. 8. Time-height cross sections of ceilometer aerosol backscatter intensity a) at Salzburg, b) at Radstadt and c) at Nötsch between April 3, 2016 12 UTC and April 7, 2016 0 UTC. The vertical white line indicates the end of the second/ the beginning of the third episode of this event.

Fig. 9. Five days integral source-receptor sensitivity fields of the lowest 2000 m above ground for the measurements at Sonnblick (a) on April 1, 2016 9 UTC to 12 UTC, (b) on April 4, 2016 6 UTC to 9 UTC and (c) on April 5, 2016 21 UTC to April 6, 2016; percentages give proportion of air prevailing in middle and southern Europe (blue box), in western Sahara and High Atlas region (green box) and in eastern Sahara (red box).

Fig. 10. Dust concentration over Europe (mg m⁻²) in the whole air column up to 50 hPa simulated with WRF-Chem for (a) April 3, 2016 12 UTC and (b) April 5, 2016 12 UTC and the contribution to the dust concentrations due to desert dust for (c) April 3, 2016 12 UTC and (d) April 5, 2016 12 UTC.

Fig. 11. Time-series of modelled PM concentrations (hourly values) within the lowest model layer within a 10 km x 10 km grid box centred at the mountain sites from the operational WRF-Chem model run, simulated PM concentrations due to wind erosion only (DUST), the proportion of dust in the modelled PM concentrations and the measured concentrations (a) at Sonnblick, (b) at Dobratsch and (c) at Feuerkogel, from March 31, 2016 0 UTC until April 9, 2016 0 UTC.
Fig. 1
Fig. 2

Daily mean PM10 concentrations depending on the flow direction.
Fig. 3
Fig. 5
Fig. 6
Fig. 7
Fig. 8
Fig. 9 (a)
Fig. 9 (c)
Fig. 10 (d)
Fig. 11 (a) Sonnblick WRF-Chem Gridbox 10 km x 10 km L0
- PM10 (model)
- DUST (model)
- % of DUST in PM10 (model)
- PM Sonnblick (observation)

Fig. 11 (b) Dobratsch WRF-Chem Gridbox 10 km x 10 km L0
- PM10 (model)
- DUST (model)
- % of DUST in PM10 (model)
- PM10 Dobratsch (observation)
Fig. 11 (c)