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Identification and monitoring of Saharan dust: An inventory representative for south Germany since 1997

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HIGHLIGHTS

- Large Ca^{2+} abundance and coarse particle volume (0.5–7 μm) indicate Saharan dust.
- The annual number of Saharan dust days shows no significant trend.
- The relative contribution of dust to total particle mass decreases from 6% to 4%.
- Sahara dust events, detected by different metrics at different stations, are consistent.
- 4 of 5 EU law threshold exceedances (daily PM10 > 50 mg/m^3) are due to Saharan dust.

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ABSTRACT

An inventory of Sahara dust (SD) events at the Hohenpeißenberg Global Atmosphere Watch station (Germany) is presented for the period 1997–2013. Based on daily in-situ measurements, high Ca^{2+}-ion concentrations and large particle volume concentrations at diameters $d_p \approx 0.5$–7 μm are inferred as indicators for days influenced by SD. The resulting SD catalogue agrees with SD time series from Schneefernerhaus, Augsburg and Jungfraujoch stations. On average, SD occurs in 5–15 SD events (SDE) per year covering about 10–60 days/yr in the mixing layer. SDE exhibit a clear seasonality with spring and early autumn maxima, and typically last for 1–3 days. SDE are equally frequent but more significant at Alpine levels due to lower background. Wet deposition of Ca^{2+} at the surface is little correlated ($R^2 = 0.14$) with particle Ca^{2+} on a daily basis and yields an average annual Ca^{2+} immission of 0.22 ± 0.04 g/m^2 yr, about 40% of which is due to SD. The majority of outstanding weekly Fe and Al depositions are associated with SDE. SD contributes about 0.5 ± 0.1 μg/m^3 to the total particle mass with a decreasing trend from 6% to 4% (−0.1%/yr) in the 1997–2013 period. Except from one, all threshold exceedances according to European legislation (daily PM_{10} > 50 μg/m^3) at Hohenpeißenberg are due to SD. Implications are discussed with respect to SD-related circulation patterns, SD-induced temperature anomalies in weather forecast models and the capability of aerosol models to capture SDE.

1. Introduction

Mineral dust significantly affects weather (Pèrez et al., 2006) and climate (Hansen et al., 1997; Philipona et al., 2009) by altering stratification and surface conditions and by its impact on the formation and droplet size of clouds. This again changes the amount (Andreae and Rosenfeld, 2008) and acidity (DeAngelis and Gaudichet, 1991; Wagenbach et al., 1996) of precipitation. The global dust radiative forcing estimation of [−0.3 to +0.1] W/m^2 has a large uncertainty, caused by the lacking knowledge of particle shape, spatial distribution and optical properties. About 80% of the forcing stems from natural sources. Mineral dust amounts to roughly 75% of the global aerosol load and about 35% of the

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primarily emitted aerosol mass (Boucher et al., 2013). Roughly half of this originates from the Sahara and is transported across the Atlantic towards South America and Eastern US or across the Mediterranean to Europe (Tegen and Schepanski, 2009; Prospero et al., 2014; Pourmand et al., 2014; Kumar et al., 2014). In Europe, Saharan outbursts are frequently observed and, decreasing towards the north, contribute 5%–30% to the total particle mass (Collaud Coen et al., 2004; Papayannis et al., 2008; Pey et al., 2013). While in southern Europe Saharan dust (SD) causes up to 20 daily threshold exceedances per year w.r.t. the European Directive 2008/50/EC, the contribution is harder to quantify for the middle and north (Pey et al., 2013). Annual mean depositions of SD range from 0.4 to 1 gm^{-2} yr^{-1} in alpine ice cores (Wagenbach and Geis, 1989; DeAngelis and Gaudichet, 1991) to several ten gm^{-2} yr^{-1} in southern Europe (Fiol et al., 2005). Dust episodes may be associated with health risks (Karanasiou et al., 2012) and dispersion of epidemic bacteria (Tobias et al., 2011), but the results are still inconsistent due to sparse data. The European regulation of PM_{10}/$PM_{2.5}$ (Linares et al., 2009), makes dust transport also a political and social-economical issue.

Varying between source regions, about 90 mass-% of SD are aluminosilicates (e.g. illite, kaolinite, montmorillonite), iron oxides (hematite, goethite) and quartz (Sokolik and Toon, 1999). The remaining <10% are mostly calcite and gypsum. Ionic Ca^{2+} and alkalinity (H^+-concentration) have been proposed as SD tracers by Wagenbach et al. (1996). K^+ and Mg^{2+} are found in SD, but also in regional minerals, smoke (K^+) and plant debris (K^+, Mg^{2+}). Besides sea-spray, volcanic ash and pollen, mineral dust is the only prominent coarse mode aerosol and thus dominates the particle volume distribution if the former can be ruled out. Dominance of illite, kaolinite and montmorillonite over hematite reflects in stronger shortwave absorption (Russell et al., 2010; Sokolik and Toon, 1999) and spectral single scattering albedo (Collaud Coen et al., 2004). Little import of mineral dust to Central Europe occurs from outside the Sahara (Küfmann, 2003; Birmili et al., 2007), but occasionally significant amounts may emerge from local soils (Goossens et al., 2001; Lequy et al., 2013) and the carbonate massif of the Northern Calcareous Alps (Küfmann, 2003). The latter is formed by the Main Dolomite, a 1–2 km thick lithostratigraphic unit made of >97% dolomite rock ($CaCO_3 \cdot MgCO_3$), which extends all along the north-eastern Alpine ridge. Eolian sediments from regional sources exhibit a clear seasonality due to snow cover in winter (Küfmann, 2003), but are not yet quantified. Thus, a regional dolomite contribution to dust also results in an association of Ca and Mg, but with a probably larger Mg portion than in SD.

The SD fingerprint is less clear in the lower mixing layer (ML) than at mountain stations or in the free troposphere (FT), where it can be observed largely unmixed by lidars and radiometry. Thus we apply a multi-factorial approach to retrieve and identify faint traces of SD in the largely variable ML aerosol in central Europe. We infer a 17-year SD inventory and compare it to independent SDE compilations at surrounding sites, based on particle composition, particle volume distribution (PVD), optical properties and Positive Matrix Factorization (PMF) analysis (section 3). We study the trend of the SD contribution to TSP (PM_{10}), the ambiguity and frequency of events and their dependence on circulation patterns (section 4). From this we draw implications for human health studies and potential impacts on weather and climate. A summary is given in section 5.

2. Experimental

2.1. Measurement sites

In-situ particle measurements are performed at the Global Atmosphere Watch (GAW) global station Hohenpeißenberg HPB (47.8°N, 11.0°E, 980 m a.s.l.) and Schneefernerhaus SFH (2650 m a.s.l., 40 km south of HPB) since 1997 and 2010, respectively and are continuously complemented. The HPB is a pre-Alpine hill, sticking out 300 m above the surrounding forest/grassland and represents rural central European conditions while SFH is located on the steep south-exposed slope 300 m below Zugspitze summit at the northern ridge of the German Alps. SFH is often above the ML, particularly in winter (Gilge et al., 2010; Flentje et al., 2010). To evaluate our approach we compare SD observations at HPB, SFH and Augsburg (AUG, 480 m a.s.l., 60 km north of HPB, ≈260,000 inhabitants). The AUG data (Pitz et al., 2011) covers the period 2005–2010 and represents lowland urban conditions. Finally, the covariance with SDE at the Swiss GAW global station Jungfraujoch (JFJ, 46.54°N, 7.98°E, 3580 m a.s.l., 250 km SW of HPB, www.ifjungo.ch) is discussed for the period 2001–2011 w.r.t. large scale dispersion and extension of SD layers, c.f. Collaud Coen et al. (2004).

2.2. Ground based in-situ observations

The size-segregated particle ionic composition is sampled since mid 1997 by a cylindrical 3-stage low-pressure Berner-Impactor BI (Wang and John, 1988), parallelled for bulk-validation by 2–3 μm pore-size quartz/teflon-filters. The samples are analysed by ion-chromatography, currently DIONEX ICS 1000 (http://www.dionex.com), after aqueous extraction (Henning et al., 2002). BI and filter probes can be analysed for SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, Na^+, Ka^+, Mg^{2+}, Ca^{2+} and H^+. The BI is equipped with pre-baked ring-like aluminium foils for each of the three stages and segregates particles into three size fractions from 6 to 1.9 μm, 1.9–0.6 μm, 0.6–0.2 μm (stage I–III), and a terminating backup which collects the smallest and previously bounced particles. The BI sampling interval, initially 24-h on weekdays and 48-h at weekends (alternating Sun/Mon changes), recently had to be reduced owing to staff limitations to 3 samples per week (01/08–04/09 and 08/11–12/13) and temporarily one sample per week (10/12–02/13). Since 2010, sampling is interrupted during fog and rain (identified by >97% RH) to avoid flooding. Daily precipitation samples are analysed by ion-chromatography and also sent to the 'Bayerisches Landesamt für Umwelt' for weekly heavy metal analyses, using Inductively Coupled Plasma-Mass Spectrometry. The inlet is PM_{10} (Digitel), except for total mass and total size distribution measurements. The sample air is heated few degrees to avoid condensation and conditioned to below 40% RH by Nafion driers. Particle size/volume distributions (dN/dlogd, dV/dlogd) are measured by optical particle sizers OPS, LAS-X (Hinds and Kraske, 1986) till 2007 and GRIMM-EDM 190 (www.grimm-aerosol.com) as of 2008. The ranges of LAS-X (0.1–7.5 μm) and EDM 190 (0.25–30 μm) both cover the relevant size range of SD particles $d_p = 0.5–7$ μm.

Particle scattering coefficients σ_{sc} are measured by an integrating 3-λ nephelometer TSI 3562 (Anderson and Ogren, 1998), the absorption coefficient σ_{abs} by a 3-λ particle soot absorption photometer PSAP (Bond et al., 1999) as of 2011 and a 7-λ aethalometer (Arnott et al., 2005) as of 2014. From these the extinction coefficient σ_{ext}, the single scattering albedo (SSA) ($\omega_0 = \sigma_{abs}/(\sigma_{abs} + \sigma_{sc})/\sigma_{ext}$) and the respective Ångström Exponents α_{sc}, α_{abs} and α_{ssa} can be calculated as measures of particle size, colour and brightness. The Ångström Exponent is defined by $\sigma_{sc} \sim \lambda^{-\alpha_{sc}}$, resulting in $\alpha_{sc} = -log(\sigma_{sc,\lambda_1}/\sigma_{sc,\lambda_2})/log(\lambda_1/\lambda_2)$ for scattering and analogous for absorption and SSA.

Quality control is performed by intercomparison of Berner with filter measurements, OPS sub-micron size spectra with Scanning Mobility Particle Sizer SMPS, and between the different photometers. The uncertainties of dV/dlogd, σ_{sc}, σ_{abs}, σ_{ext} and the SSA for the relevant SD conditions are of the order of 5–10%. Uncertainties of the chemical data depend on the ion, the sample volume,