# Aerosol characterization at the Saharan AERONET site

# 2 Tamanrasset

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#### 1 Abstract

2 More than two years of columnar atmospheric aerosol measurements (2006-2009) at 3 Tamanrasset site, in the heart of the Sahara desert, are analysed. AERONET level 2.0 data 4 were used. The KCICLO method was applied to a part of level 1.5 data series to improve the 5 quality of the results. The annual variability of aerosol optical depth (AOD) and Angstrom 6 exponent (AE) has been found to be strongly linked to the Convective Boundary Layer (CBL) 7 thermodynamic features. The dry-cool season (autumn and winter time) is characterized by a shallow CBL and very low mean turbidity (AOD~0.09 at 440 nm, AE~0.62). The wet-hot 8 9 season (spring and summer time) is dominated by high turbidity of coarse dust particles 10 (AE~0.28, AOD~0.39 at 440 nm) and a deep CBL. The aerosol-type characterization shows 11 desert mineral dust as prevailing aerosol. Both pure Saharan dust and very clear sky 12 conditions are observed depending on the season. However, several case studies indicate an anthropogenic fine mode contribution from Libya and Algeria's industrial areas. The 13 14 Concentration Weighted Trajectory (CWT) source apportionment method was used to 15 identify potential sources of air masses arriving at Tamanrasset at several heights for each 16 season. Microphysical and optical properties and precipitable water vapour were also 17 investigated.

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#### 19 **1** Introduction

The regional characterization of mineral dust, particularly close to source areas, has become a valuable tool for researchers from different fields. It will lead to reduce some uncertainties about direct radiative forcing by atmospheric aerosols that still exist (Forster et al., 2007), and to achieve a better understanding about aerosol potential impact on human health and air quality (e.g. De Longueville et al., 2010; Perez et al., 2012).

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The Sahara and its margins are the largest and most continuous dust sources in the world. Several satellite and ground based observation analysis have led to identify the base of the Ahaggar and Tibesti Mountains and the Bodélé Depression as the major sources in this area (Goudi and Middleton, 2001; Prospero et al., 2002; Ginoux et al., 2012). During the last years, several field campaigns in different locations focused on the analysis of some Saharan dust features (Todd et al., 2013, and references therein). In particular, Tamanrasset (main city in the Hoggar, also known as Ahaggar Mountains, in Algeria) hosted a specific soil and
 aerosol sampling analysis at the beginning of the 1980s (d'Almeida and Schütz, 1983), the
 African Turbidity Monitoring Network (1980-1984) for climate modelling purposes
 (d'Almeida, 1986, 1987), and the more recent African Monsoon Multidisciplinary Analysis
 (AMMA) campaign (Redelsperger et al., 2006).

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7 During AMMA intensive observing periods in 2006, Tamanrasset was a fully equipped 8 ground-based station for aerosol and radiation measurements. This campaign has provided 9 comprehensive analysis of several features at Tamanrasset and the Hoggar Mountains (e.g. 10 Flamant et al., 2007; Bou Karam et al., 2008; Cuesta et al., 2008, 2009, 2010). In addition, 11 aerosol observations carried out at Tamanrasset in 2006 have been part of selected aerosol 12 data sets used for several model validations (e.g. Toledano et al., 2009; Haustein et al., 2009, 2012; Su and Toon, 2011). In spite of these studies, limited aerosol observations, mainly 13 14 confined to shorter period campaigns, are available for this area which is strategically located 15 in the heart of the Sahara desert.

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17 Consequently, Tamanrasset was considered to be a key place to initiate the Saharan Air Layer 18 Analysis and Monitoring (SALAM) project as part of the Global Atmospheric Watch (GAW) 19 Twinning cooperation program between l'Office Nationale de la Météorologie (ONM, 20 Algeria) and the Meteorological State Agency of Spain (AEMET, formerly INM) through the Izaña GAW station (Canary Islands, Spain). In the framework of this project, at the end of 21 22 September 2006, a Cimel Sun photometer was set up at Tamanrasset and integrated into the Aerosol Robotic Network (AERONET). In 2010 the station was incorporated into the World 23 Meteorological Organization (WMO) Sand and Dust Storm Warning Advisory and 24 25 Assessment System (SDS-WAS) Regional Center for Northern Africa, Middle East and 26 Europe (http://sds-was.aemet.es/) for near-real time and long-term dust model evaluation. The 27 new aerosol dataset from Tamanrasset has been used for a preliminary characterization of 28 aerosol properties (Guirado et al., 2011), for space-based remote sensing evaluation (e.g. 29 Schuster et al., 2012), and for model validation (e.g. Tegen et al., 2013). Regarding dust optical properties, Kim et al. (2011) provide an analysis of single scattering albedo, 30 asymmetry parameter, real refractive index, and imaginary refractive index at several stations, 31 32 including Tamanrasset from 2006 to 2009.

2 The present work focuses on a detailed characterization of aerosol properties at Tamanrasset 3 site. Very preliminary results, briefly shown by Guirado et al. (2011), have been carefully 4 revised and extended taking into account corrected data. The KCICLO method has been used 5 to correct the aerosol optical depth (AOD) and the Angstrom exponent (AE) time series. 6 Specific characterizations have been made for the first time: annual evolution and seasonal 7 features of precipitable water vapour (PWV), fine mode fraction (FMF), and aerosol 8 microphysics, as well as an identification of potential source regions. The paper is structured 9 as follows: Measurement site, data sets and tools used are described in Sect. 2. In Sect. 3.1 the 10 main aerosol and PWV seasonal features are analysed, an aerosol-type classification is 11 performed and microphysical and optical properties are discussed. In Sect. 3.2 the 12 Concentration Weighted Trajectory method is used to identify potential source regions. In 13 Sect. 3.3 the transport of anthropogenic fine aerosols to Tamanrasset is discussed. In Sect. 4 14 the main concluding points are provided.

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#### 16 2 Methodology

#### 17 **2.1** Unique characteristics of Tamanrasset site

18 On 30 September 2006, a sun photometer was installed on the roof of the main building of the 19 Regional Meteorological Center (Direction Météo Régional Sud, Office National de la 20 Météorologie, Algeria) at Tamanrasset (22.79°N, 5.53°E, 1377 m a.s.l.) in southern Algeria. 21 Tamanrasset is free from industrial activities and is representative of pure desert dust aerosols 22 (Guirado et al., 2011). It is near dust sources located in Mali, southern Algeria, Libya and 23 Chad, on the northern edge of the zonal dust pathway identified by MISR (Multi-angle 24 Imaging SpectroRadiometer, onboard NASA's Terra satellite) AOD retrieval (Fig. 8). 25 Moreover, this geographical location is very significant since ground based measurements of 26 atmospheric constituents from continental Africa are very limited, especially in the 27 surrounding area of Tamanrasset. This station is involved in several international 28 measurement programs such as the Global Climate Observing System (GCOS) - Upper-Air 29 Network (GUAN), the Baseline Surface Radiation Network (BSRN), and the GAW program 30 of the WMO.

The climate of the region is modulated by the influence of the monsoon during summer and 1 2 the westerly winds during the rest of the year (Cuesta et al., 2008). In July and August 3 easterly winds, moist air masses and scarce rainfall are the prevailing weather conditions. In 4 September the influence of the westerly winds appears at high altitude and draws successively 5 closer to the ground until the end of autumn. This system is maintained, although wind strengths vary, during the winter and even springtime until June when the influence of the 6 7 easterly winds starts in layers close to the ground (Dubief, 1979). The winter season is 8 characterized by dry conditions and occasional midlevel and cirrus clouds (Cuesta et al., 9 2008).

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#### 11 **2.2 Cimel sun photometer data set**

# 12 2.2.1 AERONET data

The Cimel sun photometer (model CE-318 operating at 340, 380, 440, 500, 670, 870, 940 and 13 14 1020 nm nominal wavelengths) installed at Tamanrasset is one of the standard instruments in 15 AERONET. Data acquisition protocols, calibration procedures and data processing methods 16 are extensively described (Holben et al., 1998; Dubovik et al., 2000; Smirnov et al., 2000; O'Neill et al., 2003). Solar extinction measurements are used to compute AOD at each 17 18 wavelength, except for the 940 nm channel, used to retrieve PWV (Eck et al., 1999). AE, 19 which is a measure of the AOD spectral dependence with the wavelength of incident light, is 20 a qualitative indicator of aerosol predominant particle size and it can be computed for two or 21 more wavelengths (Schuster et al., 2006). For climatological studies, linear fit determination 22 of AE in the 440–870 nm range is computed for three or four nominal wavelengths (440 nm, 23 500 nm when available, 670 nm, and 870 nm). AERONET fine mode fraction (FMF) from the 24 Spectral Deconvolution Algorithm (SDA) (O'Neill et al., 2003) has also been included in the 25 present analysis. Furthermore, several aerosol microphysical and optical properties retrieved 26 from the AERONET inversion algorithm (Dubovik and King, 2000; Dubovik et al., 2006) are 27 discussed. Particularly, particle size distribution, volume concentration, effective radius, as well as single scattering albedo, asymmetry factor, and complex refractive index are analysed 28 29 because they are closely related to aerosol radiative effects.

The AOD uncertainty is approximately 0.01-0.02 (spectrally dependent with the higher errors 1 2 in the UV) and it alters the AE by 0.03-0.04 (Eck et al., 1999; Schuster et al., 2006). The 3 PWV uncertainty is around  $\pm 10\%$  (Holben et al., 2001). The amplitude of the errors of the 4 derived parameters from SDA retrieval varies as the inverse of the total AOD. In addition to 5 measurement errors, there are errors in the AOD retrieval due to the uncertainty in the assumed values of the spectral curvature in each mode (O'Neill et al., 2001) which are most 6 7 critical in coarse mode dominated conditions. Dubovik et al. (2002) summarized a detailed 8 description of expected error in aerosol size distribution, complex refractive index, and single 9 scattering albedo.

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11 At the present, AERONET level 2.0 at Tamanrasset is only available from October 2006 to 12 February 2009, except from 18 November 2007 to 20 June 2008. Data for the period February 2009-October 2012 will likely never be promoted to Level 2.0, and Level 1.5 data in this 13 14 period do not have the sufficient quality to be properly corrected with the KCICLO method that will be addressed below. Data after November 2012 are expected to achieve AERONET 15 16 level 2.0 and might be incorporated in the future to perform a relatively longer term analysis. Long AOD data series fulfilling the highest quality criteria are difficult to obtain in remote 17 18 stations as Tamanrasset, in which the annual exchange of instruments is difficult, and where 19 intense dust storms dirty the optics sometimes very quickly, sometimes progressively, 20 deteriorating the quality of the measurements.

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#### 22 2.2.2 KCICLO correction

The analysis of the AOD period from 18 November 2007 to 20 June 2008 reveals a systematic and strong AOD and AE fictitious diurnal cycle, most likely caused by dirtiness on the sun photometer front windows (Guirado et al., 2011). Other possible causes, such as the effect of temperature on the detector and an incorrect sun pointing, were analysed and discarded. Measurements corresponding to 340 nm and 500 nm did not achieved level 2.0 in the whole analysed period due to the significant degradation of these filters.

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The KCICLO (K is the name of a constant and "ciclo" means cycle in Spanish) method is used to detect, evaluate and correct possible calibration problems, after discarding a real

1 atmospheric effect or instrument malfunctions (Cachorro et al., 2004, 2008a). Particularly, the 2 obstruction in the optical path, due to dirtiness on the sun photometer front windows, leads to 3 a distinct and artificial diurnal cycle pattern that can be corrected using the KCICLO method. 4 This fictitious diurnal cycle is due to the systematic absolute error in the AOD measurements 5 as a consequence of calibration errors (Romero and Cuevas, 2002): the magnitude of this 6 absolute error is greatest at midday because varies as the inverse of the solar air mass 7 (Cachorro et al., 2008a). This method introduces a constant K defined as the ratio between 8 "incorrect" current and true calibration constants. K quantifies calibration factor error in such 9 a way that K=1 corresponds to correct calibration constant and K>1 (K<1) will result in an 10 overestimation (underestimation) of the current calibration constant and a convex (concave) 11 curve shape in the diurnal cycle (Cachorro et al., 2004, 2008a). AOD relative differences 12 between AERONET level 2.0 and KCICLO data series are estimated to be 8.5% (or about 13 0.01 in absolute AOD values) and 2.4% for AE (Cachorro et al., 2008b).

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15 The application of this "in situ" correction-calibration procedure requires a sufficient number 16 of clear-sky and stable days for a given period to be corrected. The selected days must fulfil a 17 set of requirements about air mass range (higher than 0.4 and typically between 1.7 and 6), 18 turbidity (AOD (440 nm) < 0.12 and variability lower than 5% in the specified air mass 19 range), number of data points (at least 12 per day), and standard deviation of the fit to 20 quantify the calibration factor error (lower than 0.01) (Cachorro et al., 2008a). Therefore, the 21 successful application of the KCICLO method over a given period is associated with a 22 sufficient number of days (5-10%) fulfilling all the above mentioned requirements. As a consequence, the application of the method is not always feasible at all stations or at all 23 24 periods of time. KCICLO method has been previously used to correct AOD data series (e.g. 25 Toledano et al., 2007; Barreto el al., 2014).

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At Tamanrasset, a sufficient number of days (94) from 18 November 2007 to 20 June 2008 were available to properly apply the KCICLO method and complete the AOD/AE data set. This method confirmed a calibration shift between November 2007 and June 2008. Only two different correction periods, i.e. two different types of lenses contamination (amount of dirtiness and lenses affected), were detected and the corresponding mean K values (Table 1) were computed. A part of original and corrected AOD and AE data for both periods is shown 1 in Fig. 1. Note that the fictitious diurnal cycle is largely reduced both in the AOD and the

2 derived AE.

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4 Additionally, it was possible to apply an external quality control of the KCICLO correction. 5 Since 1995, in the framework of the GAW program, a J-309 hand-held sun photometer 6 supplied by the National Oceanic and Atmospheric Administration (NOAA) (Reddy, 1986) 7 has been operated at Tamanrasset. The photometer is characterized by a 2.5° full angle field of 8 view and two 10nm-bandwidth filters centred at 386 and 506 nm, respectively. AOD 9 measurements at 500 nm taken at 9, 12 and 15 UTC were used in this work. Data from 10 October 2006 to February 2009 were compared to the closer time AERONET measurements 11 at 440 nm (±15 minutes as time coincident criterion). AOD measurement scatter plot between 12 NOAA and three AERONET data sets is shown in Fig. 2 and the corresponding linear regression parameters are provided in Table 2. After applying the KCICLO correction the 13 14 correlation coefficient increases to 0.981 for this period (0.968 before correction).

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#### 16 **2.2.3 Time series**

Following the data processing and quality control procedures described above, AERONET 17 18 level 2.0 and KCICLO-corrected level 1.5 data (AOD and AE) were used for aerosol characterization. Due to the degradation of the 500 nm filter, AOD measurements at 440 nm 19 were selected for analysis. However, since AOD at 500 nm is more suitable for satellite and 20 21 modelling comparisons, it was estimated from AOD (440 nm) and AE (440-670-870 nm) 22 applying the Angstrom power law (Ångström, 1929). With regard to the PWV record, 23 AERONET level 1.5 measurements were not affected by any fictitious diurnal cycle. The water vapour optical depth, and consequently the PWV product, is not strongly affected by 24 25 obstructions in the optical path because the calculation algorithm is based in a subtraction of experimental measurements (Schmid et al., 2001). Therefore, the analysed PWV data series 26 27 comprised AERONET levels 2.0 and 1.5 when level 2.0 is not available. Limitations and 28 special features regarding the analysed AERONET inversion retrievals for single scattering 29 albedo and complex refractive index will be discussed in Sect. 3.1.4. All the analysed daily, 30 monthly, and seasonal averages have been calculated from the corresponding sun photometer 31 single measurements.

## 1 2.3 Ancillary data

# 2 2.3.1 Meteorological radiosonde data

3 A GCOS-GUAN meteorological radiosonde (Vaisala RS92) is launched twice a day (at 00 UTC and 12 UTC) at Tamanrasset airport: data available at the University of Wyoming web 4 5 site (http://weather.uwyo.edu/upperair/sounding.html). Radiosonde data at 12 UTC were used 6 for calculation of the Convective Boundary Layer (CBL) top altitude from 2006 to 2009. The 7 criteria used to account for the overshooting thermals have been  $\Delta\theta/\Delta z \ge 0.0025$  K/m and 8  $\theta_{top} - \theta_{base} \ge 1$  K, where  $\Delta \theta / \Delta z$  is the potential temperature lapse rate and  $\theta_{top}$  and  $\theta_{base}$  refer to 9 the top and base of the layer, respectively (Heffter, 1980; Cuesta et al., 2008). Additionally, 10 PWV retrieved from radiosonde was compared with corresponding AERONET PWV as it will be shown in Sect. 3.1.5. Estimated PWV precision of the radiosonde RS92 is around 5% 11 12 but for very dry conditions it is about 10-20% (Miloshevich et al., 2009).

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# 14 **2.3.2** Aerosol extinction vertical profiles

15 The Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) is an elastic-backscatter lidar on-board the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation 16 (CALIPSO). CALIOP emits linearly polarized light at 532 and 1064 nm to provide vertically 17 resolved observations of aerosols and clouds on a global scale (Hunt et al., 2009; Winker et 18 19 al., 2009). Aerosol extinction features at certain heights have being identified using CALIOP level 2 version 3.01 extinction profiles at 532 nm over Tamanrasset (within a 1.5° radius) 20 21 with a vertical resolution of 60 m (below 20.2 km height) and a horizontal resolution of 5 km. 22 Data from the period 2007-2008, downloaded from NASA database 23 (https://eosweb.larc.nasa.gov/cgi-bin/searchTool.cgi?Dataset=CAL IIR L1-Prov-V1-10),

have been filtered following the methodology of Tesche et al. (2013).

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# 26 **2.3.3 Concentration Weighted Trajectory**

Concentration Weighted Trajectory (CWT) source apportionment method (Seibert et al.,
1994; Hsu et al., 2003) was used to identify pathways of aerosol laden air masses for the
period 2006-2009 in the dry season (from November to February) and the wet season (from
April to September). The resulting information about air mass pathways was combined with

the information about aerosol source regions reported by several authors (Sect. 3.2 and Fig. 8) 1 2 to detect potential sources affecting Tamanrasset. This method combines data measured at the 3 receptor site with air mass back trajectories. Although this method was originally designed 4 and widely used for weighting trajectories with concentrations measured at a receptor site, we 5 used AERONET daily AOD and AE observations at Tamanrasset to identify aerosol content 6 and type respectively. A similar approach to connect distinct sources with different aerosol 7 types has been previously performed by other authors (e.g. Naseema Beegum et al., 2012). A 8 weighted AOD or AE value is assigned to each grid cell by averaging the values associated 9 with the trajectories crossing that grid cell:

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$$C_{ij} = \left(\sum_{k=1}^{N} n_{ijk}\right)^{-1} \cdot \sum_{k=1}^{N} C_k n_{ijk}$$
 (1)

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where  $C_{ij}$  is the averaged weighted AOD or AE value in the (i,j) grid cell,  $C_k$  is the AOD or AE value observed at the receptor point on arrival of *k*th-trajectory, *N* is the total number of trajectory end-points in the (i,j) grid, and  $n_{ijk}$  is the number of *k*th-trajectory end-points in the (i,j) grid cell, i.e., the time spent in the *ij*th-cell by the *k*th-trajectory. The denominator corresponds to (i,j) grid cell number density. In order to reduce the uncertainty caused by cells with few trajectory end-points, an arbitrary weight function  $W_{ij}$  (Polissar et al., 1999) was applied:

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$$W_{ij} = \begin{cases} 1.00 & 80 < n_{ij} \\ 0.70 & 20 < n_{ij} \le 80 \\ 0.42 & 10 < n_{ij} \le 20 \\ 0.05 & n_{ij} \le 10 \end{cases}$$
(2)

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Three-dimensional 5-day back trajectories were calculated with a one-hour time resolution using the Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) version 4.0 (Draxler and Hess, 1998). The end-point was set at Tamanrasset (22.790°N, 5.530°E), and back trajectories were calculated at ground level, 2600 and 5600 m above ground level (a.g.l.) for each day in the period 2006-2009 at 12 UTC with wind fields from the GDAS
 meteorological data set. The vertical model velocity was taken into account.

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The  $C_{ij}*W_{ij}$  values in the geographical domain long= [30°W, 30°E], lat = [5°N, 50°N] were mapped separately for the dry and the wet seasons and for back trajectories ending at the three levels mentioned above. These maps were examined to identify potential source areas or pathways of polluted air masses. The CWT method is able to distinguish major sources from moderate ones (Hsu et al., 2003).

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10 Besides CWT analysis, Potential Source Contribution Function (PSCF) maps (Ashbaugh et 11 al., 1985) were also obtained in order to identify the direction and sources of air masses 12 causing high AOD and AE values at Tamanrasset. The PSCF method estimates the conditional probability of each pixel of the geographical domain being a source location, 13 14 using back trajectories arriving at the study site. The results are plotted on a map describing the spatial distribution of potential source regions. We used the same back trajectories, AOD 15 and AE values and arbitrary weight function, Eq. (2), for both the PSCF and CWT methods. 16 17 Our resulting PSCF maps are in good agreement with CWT ones. We only show CWT results 18 because they provided the same information on potential sources location plus additional 19 information on the intensity of the sources, as already mentioned.

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#### 21 **3** Results and discussion

#### 22 **3.1 Characterization**

# 23 **3.1.1 Temporal evolution and statistics**

From October 2006 to February 2009, a total of 31,800 cloud-free valid AOD observations from 790 days (92% of the days in the period) are available. After KCICLO correction, AOD and AE values are globally lower (around 8% and 17% respectively) than the time series shown by Guirado et al. (2011). The AOD, AE and FMF monthly statistics are presented in Figs. 3a, 3b, and 3c, respectively. AOD remains stable around 0.1 from November to February (absolute minimum of 0.07 in January). Conversely, AOD exceeds 0.3 from April to

September reaching an absolute maximum of 0.43 in June (Fig. 3a and Table 3). High AOD 1 2 variability (standard deviation>0.30) is observed for high monthly AOD records (from April 3 to August except May) while the lower variability (STD≈0.10) coincides with the lower AOD 4 observations (from November to January). March and October have been considered 5 transition months between the main two seasons because they alternately show similarities to each season. Similar results were reported by Kim et al. (2011) from a different approach. 6 7 Their analysis was limited to "dust aerosol" properties by selecting data with large AOD 8  $(\geq 0.4)$  and very low AE  $(\leq 0.2)$ . According to these criteria, non-dust aerosols were identified 9 from November to February at Tamanrasset station.

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The annual cycle of AE and FMF is the opposite of AOD (Fig. 3). The dry season is characterized by higher AE and FMF values, reaching a maximum in January (0.69 and 0.58, respectively) and December (0.72 and 0.57, respectively) and decreasing until May (minimum of 0.15 and 0.24, respectively). A secondary maximum is observed in August with AE and FMF values of 0.44 and 0.38, respectively, associated with a decrease of the coarse mode and a slight increase of the fine mode. Such increase will be analysed in detail in Sect. 3.3.

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18 Concerning the pattern shown in Fig. 3, Cuesta et al. (2008) identified a marked seasonal 19 evolution of atmospheric aerosol content and its optical properties linked to the monsoon 20 regime throughout 2006. Guirado et al. (2011) stated the clear and opposite seasonal cycle of 21 AOD and AE, compared them with the CBL, and defined a dry-cool season (autumn and 22 winter) and a wet-hot season (spring and summer). The CBL, PWV, and corrected AOD and AE time series are presented in Fig 4. Daily mean AOD at 500 nm was estimated (not shown 23 24 for the sake of brevity). Relative differences between AOD at 500 nm and 440 nm were 25 mainly below 0.01, except for AOD values above 0.1 that were sometimes higher (0.04 as 26 maximum). The dry-cool season is characterized by low AOD (~0.09 at 440 nm), not very low AE values (~0.62) and low PWV (~0.51 cm). The wet-hot season is characterized by 27 28 higher mean AOD (~0.39), lower AE (~0.28), and double the autumn-winter time PWV 29 values (~1.06 cm). A statistical summary of the data series is given in Table 4.

A strong and thick CBL drives the wet season (Fig. 4a). The properties of the transported air 1 2 masses are a part of the atmospheric phenomena that have an influence on the evolution of the 3 CBL height throughout the year (Cuesta et al., 2008). Moreover, this evolution is linked to the 4 seasonal climatic features at Tamanrasset, described at the end of Sect. 2.1. The wet season, 5 affected by the monsoon regime, is characterized by strong and frequent mineral dust storms (Guirado et al., 2011) when the deep CBL favours the vertical mixing of lifted dust layers 6 7 (Cuesta et al., 2009). In this period, the fully developed CBL (4-6 km a.g.l.) coincides with 8 the higher AOD and PWV records at Tamanrasset. On the contrary, during the rest of the year 9 the prevailing dry westerly flow leads to a shallow CBL (1-2 km a.g.l.) with lower AOD and 10 PWV records. These results are in agreement with Cuesta et al. (2008), who reported a 11 summer season driving by a 5 to 6 km deep layer which evolved from a 1.5 to 2 km shallow 12 layer in winter during 2006. In addition, in August and September 2006 water vapour mixing 13 ratio doubled dry winter season records.

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Guirado et al. (2011) showed overall frequency histograms of AOD and AE. Due to the 15 16 observed seasonal pattern, frequency distributions of AOD and AE for the dry and wet seasons are shown in Fig. 5. AOD shows a unimodal positively skewed distribution for both 17 18 seasons. The wet season modal value is 0.15 (but only 35% of data below 0.15) while dry 19 season mode is narrower (90% of data are below 0.15) and centred in 0.1. These features lead 20 to a wider distribution for the whole data set, centred in 0.1 and showing a 60% of AOD data below 0.15 (Guirado et al., 2011), what indicates a cleaner atmosphere than sites located in 21 22 the Sahel where about 85% of the AOD values are above 0.15 (Basart et al., 2009). This could 23 be partly explained by the station height. On the contrary, AE shows a bimodal distribution 24 for both seasons. The dry season distribution is slightly bimodal (0.4 and 0.7 modal values) and symmetrical (mean and median AE are equal as it can be seen in Table 4). Whereas the 25 26 AE in the wet season distribution is positively skewed showing a narrowed first mode centred at 0.15 and a less pronounced but wider second mode centred at 0.4 (which coincides with the 27 modal value of the first mode of the dry season). 28

#### **3.1.2 Aerosol classification**

2 Guirado et al. (2011) used the graphical method proposed by Gobbi et al. (2007) to identify 3 aerosol types at Tamanrasset. This method relies on the combined analysis of AE (440-870 4 nm) and its spectral curvature, represented by the Angstrom exponent difference 5  $\delta AE = AE(440-670 \text{ nm}) - AE(670-870 \text{ nm})$ . These coordinates are linked to FMF (%) and aerosol fine mode size (µm) (Fig. 6) by reference points corresponding to bimodal size 6 7 distributions of spherical particles which have been determined using the Mie theory on the 8 basis of typical refractive index of urban/industrial aerosol (m=1.4-0.001i). The assumption of 9 spherical particles is not expected to impact significantly on the results (Gobbi et al., 2007). 10 Moreover, the level of uncertainty of this graphical method is of the order of  $\pm 25\%$  for aerosol 11 fine mode radius ( $R_f$ ) and  $\pm 10\%$  for FMF computed for refractive index varying between m=1.33-0.000i (typical of water droplets) and m=1.53-0.003i (typical of mineral dust 12 aerosols). This method was applied to AERONET level 2.0 observations which verify 13 14 AOD>0.15. This limit was selected in order to avoid errors larger than  $\sim$ 30% in AE and  $\delta$ AE, 15 as advised by Gobbi et al. (2007). Basart et al. (2009) applied this graphical methodology to 16 track and characterize mixtures of pollution and mineral dust confirming the robustness of the 17 method. Since ~95% of AOD observations during the dry season are below 0.15 (Fig. 5a), the 18 graphical method performed only for this period would not be representative. Thus, the same 19 graph shown by Guirado et al. (2011), corresponding to the whole data set, was analysed.

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21 The aerosol features at Tamanrasset (Fig. 6) are similar to those found at other arid and desert 22 areas, such as Banizombou or Saada, reported by Basart et al. (2009). Large variations of 23 AOD with AE almost inversely proportional to AOD are shown, thus higher extinctions are 24 linked to larger particles. In addition,  $\delta AE$  is negative or slightly positive indicating a large 25 dominance of one-particle mode. Typical pure Saharan dust conditions (red rectangle in Fig. 26 6) are characterized by high-extinction values (AOD>0.7) with AE<0.3 and  $\delta$ AE<0 that 27 corresponds to FMF<40% and Rf~0.3µm. Aerosols presenting higher AOD (up to 0.4) than 28 expected for AE values ranging between 0.6 and 1.1 are observed in 8.7% of the cases (green 29 rectangle in Fig. 6). They are characterized by variable  $\delta AE$ , FMF and Rf ranging between -30 0.3 and 0.2, 30% and 70%, and 0.10 µm and 0.20 µm, respectively. This pattern can be 31 associated with a mixture of mineral dust and smaller particles of another origin (Basart et al., 2009), and it is observed during summer. Biomass burning fine particles are discarded 32

because they are emitted in winter time in the Sahel region. Thus fine particles may have an
 urban or industrial origin as indicated by Guirado et al. (2011). This will be discussed in Sect.
 3.3.

4

# 5 3.1.3 Aerosol microphysics

6 Multi-annual monthly means of particle size distribution and volume concentration have been 7 analysed for the period 2006-2009 (Fig. 7a and Table 5). A slight bimodality is observed with 8 a strong predominance of coarse mode and a quite stable coarse modal geometrical radius 9 throughout the year with values around 2.24 µm. This value is within the radius interval (1- $3.5 \mu$ m) of maximum aerosol volume distributions showed in most of the aircraft campaigns 10 performed in central Sahara and compared by Ryder et al. (2013). At 2.24 µm, coarse mode 11 volume concentration is lower during the dry season (~0.03  $\mu$ m<sup>3</sup>  $\mu$ m<sup>-2</sup> in December), when 12 minimum AOD values are recorded, and then starts to grow peaking in July (~ $0.25 \,\mu\text{m}^3 \,\mu\text{m}^{-2}$ ). 13 Standard deviations are of the same order as mean values (Table 5) indicating high variability 14 15 of daily measurements. Fine mode concentration shows the same seasonal pattern as coarse mode but with values decreased by a factor of  $\sim 10$  (wet season) and  $\sim 6$  (dry season). The 16 17 presence of both submicron and coarse modes throughout the year was also observed by 18 Cuesta et al. (2008) through the analysis of in situ aerosol size distributions at Tamanrasset in 19 2006. They reported variability between the two modes lower than 10 to 15% regardless of 20 the season.

21

Daily fine mode volume fraction ( $V_{f}/V_{t}$ ) ranges between 0.03 and 0.46 (Fig. 7b) showing the 22 23 dominance of coarse mode. However, as it was discussed about FMF in Sects. 3.1.1 and 3.1.2, 24 fine or coarse particles dominate the contribution to total AOD depending on the season. The 25 relationship between these two fine mode quantitative parameters is shown in Fig. 7b. During the dry season, FMF and  $V_f/V_t$  are roughly linearly distributed. During the wet season, coarse 26 27 particles dominate in terms of both optical depth and volume concentration. However, few 28 measurements meeting FMF>0.5 and  $V_f/V_t>0.25$  are found in the wet season and most of them are linked to the fine aerosol presence analysed in Sect. 3.3. 29

1 Total effective radius follows an expected opposite seasonal pattern to AE, showing (Table 5) 2 a maximum in May (0.86 µm), a minimum in November-December (~0.58 µm) and a secondary minimum in August (0.61 µm). Regarding fine mode effective radius, it reaches a 3 4 maximum during the dry season (~0.16 µm in January and December) decreasing toward the lowest values in July and August (~0.12 µm), a seasonal trend close to the opposite of AOD. 5 6 Similarly, coarse mode effective radius show the highest mean value in January (1.92 µm) 7 and appears to be almost stable during the wet season ranging between 1.62  $\mu$ m and 1.72  $\mu$ m. 8 This last result, coarse mode effective radius decreasing for higher coarse mode concentrations under desert dust conditions (Table 5), has been previously reported and linked 9 10 to a practically monomodal volume particle size distribution (e.g. Prats et al., 2011, and 11 references therein).

12

# 13 **3.1.4 Aerosol optical properties**

14 AERONET level 2.0 retrievals for single scattering albedo (SSA) and complex refractive 15 index are limited to measurements of AOD (440 nm) > 0.4. The reason is that the accuracy of these two parameters significantly decreases under lower aerosol loading conditions: 80-16 17 100% and 0.05-0.07 for real and imaginary part of refractive index, respectively, and 0.05 for 18 SSA (Dubovik et al., 2000, 2002). Therefore, no information of these parameters is available 19 in the AERONET database for the dry season at Tamanrasset. Regarding the wet season, dust 20 optical properties (from March to October) are reported by Kim et al. (2011). To perform an 21 analysis for the dry season, we have filtered Level 1.5 data following the same AERONET criteria but applying a smaller threshold to AOD (above 0.1, instead of 0.4, at 440 nm). A 22 23 similar approach has been previously considered by other authors to investigate the role of fine aerosols on the absorption (e.g. Mallet et al., 2013). 24

Imaginary part of refractive index, SSA, and asymmetry parameter are relatively constant in the interval 675-1020 nm during both the dry and the wet seasons (Table 6). However, SSA is lower at 440 nm whereas both imaginary part (absorption) of refractive index, as well as asymmetry parameter, are higher at 440 nm. These spectral patterns of SSA and complex refractive index agree with dust dominance conditions (e.g. Dubovik et al., 2002). For the dry season this spectral dependency is smoothed and asymmetry parameter is slightly lower due to the decrease in the coarse mode dominance. Nevertheless, no substantial differences are found at overall aerosol optical properties between the dry and the wet season. It is worth noting that real part of refractive index ranges between 1.43 and 1.46 during the wet season. These values are lower than expected for dust conditions. Deviations ranging ± 0.05 or more from 1.53 have been previously reported by Dubovik et al. (2002) and references therein.

5

# 6 3.1.5 Precipitable water vapour

7 The observed PWV atmospheric content shows an annual cycle quite similar to that of the 8 CBL (Figs. 4a and 4b). The lowest multi-annual monthly mean of PWV (Table 3) is observed 9 in January ( $0.37 \pm 0.16$  cm) showing a low year to year variability and increases during winter and spring peaking in August (1.39  $\pm$  0.45 cm) under the monsoon regime. PWV retrieved 10 11 from radiosondes launched at 12 UTC have been compared (not shown) with the 12 corresponding AERONET PWV (average of the measurements taken from 12 UTC to 13 13 UTC), observing a good correlation (0.94) for an overall number of 610 coincident 14 measurements. The slope of the least-squares regression line is 1.14 and the RMS error is 1.15 15 mm. These results are in good agreement with similar comparisons, such as that performed by Schneider et al. (2010), who reported a correlation of 0.96 between 675 AERONET and 16 17 radiosonde coincident measurements (one hour as temporal coincidence criterion) at Izaña 18 station.

19

#### 20 **3.2** Potential source regions

#### **3.2.1 Concentration Weighted Trajectory analysis**

Recently, several comprehensive reviews of potential dust sources in Northern Africa (e.g. Formenti et al., 2011; Ginoux et al., 2012) have been provided. However, our goal is to identify the potential dust sources affecting Tamanrasset station. This study has been performed through the analysis of primary air mass pathways and their relationship with AERONET AOD and AE measurements at Tamanrasset.

27

CALIOP aerosol extinction profiles at 532 nm (Figs. 8c and 8d) have been analysed to link
aerosol extinctions and air mass pathways at certain heights. The 20th percentile of the

extinction in the wet season (Fig. 8d) has been selected as threshold of pristine conditions. 1 2 The CBL top features identified from CALIOP agree quite well with that obtained from the 3 radiosondes. Taking into account the averaged CALIOP profiles, HYSPLIT back-trajectories 4 at several heights have been calculated for each day of the period 2007-2008. The end-point 5 heights of the back-trajectories have been selected according to the CBL top height during both the dry and the wet seasons. The three selected height levels provide information about 6 7 air mass transport near surface (ground level), at an intermediate layer (2600 m a.g.l.), which 8 is just above the CBL top in the dry season and within the CBL during the wet season, as well 9 as at 5600 m a.g.l., above the CBL (free troposphere) all year long (Figs. 4a, 8c and 8d). A 10 first cluster analysis was performed using the k-means clustering algorithm following Jakob 11 and Tselioudis (2003) procedure. However, no-conclusive results were found due to the 12 variability of the cluster classification obtained for each season (dry and wet) and for each 13 altitude. For this reason CWT method was applied to AOD and AE parameters.

14

15 Air mass back trajectories at 2600 and 5600 m a.g.l. show a clear westerly component in the 16 dry season (Figs. 9c and 9d), driven by the general circulation, since these levels correspond 17 to free troposphere over the relatively low CBL top. The dry season is characterized by low 18 AOD and rather high AE associated with short air mass back-trajectories at ground level from 19 the first quadrant (Figs. 9a and 9b). Dust source regions identified as 1 and 2 in Fig. 8 might 20 potentially affect Tamanrasset in this season. The region located in the triangle formed by 21 Adrar des Ifoghas, Hoggar Mountains and Aïr Massif (dust source 1, Fig. 8) has been 22 previously identified (d'Almeida, 1986; Prospero, 2002; Schepanski et al., 2009; Alonso-23 Pérez et al., 2012) as a Saharan dust source formed by a drainage system of ephemeral rivers 24 and streams. This source is sensitive to the effects of mesoscale winds intensified by the orography (Ginoux et al., 2012). A second potential dust source (dust source 2, Fig. 8) 25 26 extends from the northwest side of the Tibesti Mountains in Chad over the eastern Libyan 27 Desert (d'Almeida, 1986; Caquineau et al., 2002; Prospero, 2002; Ginoux et al., 2012). This 28 source is formed by a large basin with sand seas and the northern part is marked with a chain 29 of wadis (and associated complexes of salt/dry lakes). It is active during much of the year but 30 it is especially intense in May–June.

In relation to AE (Fig. 9b), the highest values (smaller particles) are found around Tamanrasset. It could be the result of a mixture of desert dust and local pollution produced by cooking and heating bonfires that use firewood, common in this region, which are not well dispersed by the low-level atmospheric circulation. The potential influence of biomass burning from the Sahel region to Tamanrasset during the dry season can be considered as inexistent according to the CWT analysis.

7

8 In the wet season, only the 5600 m a.g.l. level is over the top of a high CBL typical of 9 summertime, showing mainly westerly trajectories (Fig. 10e). In fact, CWT analysis for AOD 10 and AE at ground and 2600 m a.g.l. levels give similar results suggesting a well-mixed CBL 11 in this season. AOD and AE CWT plots at ground and 2600 m a.g.l. (Figs. 10a, 10b, 10c, and 12 10d) clearly show a curved dust pathway with relatively high values of AE (smaller particles) 13 from northern Central Libya passing over dust sources 1 and 2 as occurs in the dry season. A 14 second curved dust pathway from the Libyan-Tunisian border (Caquineau et al., 2002) (dust 15 source 5, Fig. 8) is observed to transport larger particles (low AE) to Tamanrasset at ground 16 level. A few air mass trajectories originate from the west passing over the large dust source 4 17 (Fig. 8) located in northern Mali, northern Mauritania and the western flanks of Hoggar 18 Mountains (Prospero, 2002; Brooks and Legrand, 2003, Alonso-Pérez et al., 2012). It is a 19 complex distribution of dust sources marked with extensive dune systems which is a 20 particularly active source from April to September.

21

Regarding one of the most significant dust sources in the world, the Bodélé Depression
(Goudi and Middleton, 2001; Prospero, 2002; Brooks and Legrand, 2003) (dust source 3, Fig.
8), CWT analysis shows that it is a minor dust source affecting Tamanrasset.

25

# 26 **3.2.2 Mesoscale Convective System analysis**

Mesoscale weather systems (dry boundary layer convection, "haboob" dust storms, nocturnal
low-level jets, and southerly monsoon flow) influence dust emission, transport, and deposition
over Central Western Sahara (Marsham et al., 2008, 2013; Knippertz and Todd, 2010, 2012;
Ashpole and Washington, 2013). However, Mesoscale Convective Systems (MCSs) cannot be
well captured by global meteorological models or regional dust models (Marsham et al.,

2011; Heinold et al., 2013) as well as by HYSPLIT back-trajectory parameterization. During
 2006, Cuesta et al. (2008) observed several summertime dust transport events over
 Tamanrasset associated with MCSs. Therefore, we have performed an additional analysis of
 that based on HYSPLIT back-trajectories to identify the influence of MCSs.

5

6 We have analyzed 21 episodes of MCSs that have been selected through comparison between 7 observed AERONET AOD and NMMB/BSC-Dust model AOD over Tamanrasset in the 8 period 2007-2008 (Fig. 11). The simulation of the Non-hydrostatic Multiscale Model 9 (NMMB) Barcelona Supercomputing Center (BSC) v1 is generated using the National Center 10 for Environmental Prediction (NCEP) reanalysis-II (1° grid) and initial and boundary 11 conditions from the Global Land Data Assimilation System (GLDAS). The resolution is set to 12 at 0.5° in the horizontal and to 40 hybrid sigma-pressure model layers. A detailed description of the model is provided by Pérez et al., 2011. NMMB/BSC-Dust model properly reproduces 13 14 dust transport associated with synoptic-scale meteorological processes observed during most part of the year (Fig. 11). However, from June to September, although the AOD trend is well 15 16 reproduced, the model is not capable to capture strong and fast dust outbreaks associated with 17 MCSs. The summertime observation-model AOD discrepancies have been used to identify 18 the potential MCSs affecting Tamanrasset. The convective origin of each event has been 19 evaluated by using high temporal and spatial RGB dust composites from Meteosat Second 20 Generation-Spinning Enhanced Visible and Infrared Imager (MSG-SEVIRI) sensor combined 21 with European Centre for Medium-range Weather Forecasts (ECMWF) ERA-Interim 22 reanalysis data from IFS-Cy31r model analysis. Satellite information and meteorological data 23 were jointly computed and visualized with McIDAS (Man computer Interactive Data Access 24 System) software.

25

Once identified and confirmed all the MCS events impacting Tamanrasset, the Moderate Resolution Imaging Spectroradiometers (MODIS) Deep Blue 550 nm AOD retrieval has been used in a similar approach to Roberts (2014) and Roberts et al. (2014). The advantage of MODIS Deep Blue aerosol retrieval algorithm regarding other satellite products over bright surfaces in the visible (such as deserts) is that the former employs radiances from the blue channels where the surface reflectance is relatively low (Hsu et al., 2004; 2006). The MODIS Deep Blue composite AOD and AOD anomaly (calculated over the 2007-2008 summertime

mean value) have been analyzed to identify dust uplift sources associated with the 21 daily 1 episodes of maximum AOD driven by MCS events (Fig. 12).

- 2
- 3

4 Several regions with high AOD, including the surrounding area of Tamanrasset, are shown in 5 the MODIS Deep Blue averaged AOD map (Fig 12a). However, a strong positive AOD anomaly (above 0.20) is only shown south Tamanrasset (Fig 12b), matching with dust source 6 1 and surroundings (Fig. 8), as a consequence of the presence of MCSs in this area modulated 7 8 by northward displacement of the intertropical discontinuity (ITD). The HYSPLIT back-9 trajectories show that air flow getting Tamanrasset during these events comes from the 10 positive AOD anomaly region south of Tamanrasset. Simultaneously, a negative AOD 11 anomaly observed over eastern Mali is probably caused by rainfall associated with MCSs, 12 since on previous days to those in which a model-observation AOD anomaly is observed, the negative AOD anomaly is located to the east, south Tamanrasset (Fig 12c). These results are 13 14 in good agreement with Roberts (2014) and Roberts et al. (2014) who analyzed 31 15 anomalously rainy episodes in the Sahara and northern Sahel linked to dust uplift in the area.

16

#### Case study: anthropogenic aerosols 17 3.3

18 Some evidences of the arrival of fine particles to Tamanrasset during summer have been 19 observed in agreement with Cuesta et al. (2008) and Guirado et al. (2011). The former 20 reported a small but non-negligible contribution of fine particles to the total AOD throughout 2006. The multi-annual monthly means of AE and FMF (Fig. 3) show a local maximum in 21 22 August, i.e. a decrease of the coarse mode and a slight increase of the fine mode. In addition, 23 a mixture of fine aerosols and mineral dust has been identified mainly in July, August and 24 September (Fig. 6). The potential sources of these fine particles are indicated by the CWT 25 maps for AE (Figs. 10b and 10d) showing smaller particles arriving to Tamanrasset primarily from Central Libya through a well-defined transport pathway. 26

27

28 Frequent mixture of particulate pollutants with desert dust in the Saharan Air Layer (SAL) has 29 been reported by Rodríguez et al. (2011). In Izaña GAW observatory (Tenerife), they 30 observed that dust exported from North Africa to the North Atlantic was mixed with fine

nitrate and ammonium sulphate particles linked to emissions in oil refineries and power plants 1 2 of Algeria, Morocco and Tunisia. The CWT maps (Fig. 10) indicate Libya and Algeria as sources of the pollutants affecting Tamanrasset. Industrial activities in these countries have 3 4 been identified using the Defense Meteorological Satellite Program (DMSP) Nighttime Lights (Elvidge et al., 1997). The DMSP Operational Linescan System (OLS) has the capability to 5 6 derive Nighttime Lights of the World data sets and distinguish four primary types of lights: 7 human settlements such as cities, towns, and villages (white), fires (red), gas flares (green), 8 and heavily lit fishing boats (blue). Green light areas (Fig. 13) identified the location of gas 9 flares (i.e. oil wells, refineries, or chemical plants) in Algeria and Libya.

10

The residence time index (Alonso-Pérez et al., 2007) accounts for the percentage of time that an air parcel remained over a horizontal grid cell defined in a geographical domain before reaching a receptor site at a predefined altitude range. This index has been used to select several case studies of fine aerosol transport to Tamanrasset from some regions of Libya and Algeria, as suggested by Guirado et al. (2011). Residence time has been computed for these predefined regions from 5-day HYSPLIT back-trajectories at ground level and 2600 m a.g.l. end point altitudes.

18

19 Nine days in July, August and September 2007 and August 2008 characterized by daily mean 20 AE above 0.70 have been displayed in Fig. 13. Most of the trajectories both at ground level and 2600 m a.g.l. cross the western part of the northern Libyan gas flare zone and the 21 22 industries located in the southwest. These trajectories are up to 32% of time over the predefined Libyan zone. They are characterized by higher AE (~0.90) than the average 23 24 corresponding to the wet season (AE~0.28). On 29 August 2008 the back-trajectory arriving 25 at Tamanrasset at 2600 m a.g.l. shows air mass transport over the Algerian gas flares with 26 AE~0.73 (Fig. 13). It should be noted that optical properties of anthropogenic aerosols show 27 significant variability depending on different factors (Dubovik et al., 2002). In spite of this, 28 available filtered level 1.5 optical properties for the nine events have been analysed (not 29 shown). Two different patterns have been identified. On the one hand, a slight decrease in SSA and smaller differences between SSA at 440 nm and 675-1020 nm interval have been 30 observed, indicating the presence of other absorbing particles apart from dust, such as organic 31 32 or elemental carbon. On the other hand, several events in August 2008 show slight SSA

spectral dependency and values around ~0.96 (whereas August mean value is around 0.89)
 indicating the presence of sulphate and/or nitrate aerosols. These results agree with Rodríguez
 et al. (2011) observations.

4

#### 5 4 Summary and conclusions

6 Tamanrasset is a strategic site for aerosol research placed in the heart of the Sahara desert. An 7 aerosol characterization at this site has been provided based on more than two years (October 8 2006 to February 2009) of AERONET level 2.0 and KCICLO-corrected Cimel sun 9 photometer measurements. The top of the Convective Boundary Layer (CBL) over 10 Tamanrasset has been characterized by both radiosonde data and CALIOP extinction vertical 11 profiles. A strong seasonal cycle linked to the CBL is observed. The dry-cool season (November-February) is characterized by a shallow CBL, low aerosol optical depth (AOD) 12 13 (~0.09 at 440 nm), moderate-low Angstrom exponent (AE) values (~0.62) and low 14 precipitable water vapour (PWV) (~0.51 cm). The wet-hot season (April-September) is 15 characterized by a deep CBL, higher AOD (~0.39 at 440 nm), low AE (~0.28) and higher PWV (~1.06 cm) and it is affected by strong and frequent dust storms. March and October are 16 17 considered transition months. The AOD shows the same but opposite seasonal cycle to the AE and fine mode fraction (FMF). AOD remains stable around 0.1 from November to 18 19 February but exceeds 0.3 from April to September, reaching an absolute maximum of 0.43 in June. The maximum AE and FMF observations are reached in December (0.72 and 0.57, 20 21 respectively) and January (0.69 and 0.58, respectively), tending to decrease until May 22 (minimum of 0.15 and 0.25, respectively). Minimum PWV is recorded in January (0.37  $\pm$ 23 0.16 cm) whereas maximum values are reached in August (1.39  $\pm$  0.45 cm) linked to the 24 monsoon regime.

25

Coarse mode (modal radius around 2.24  $\mu$ m) prevails over the fine mode (modal radius around 0.10  $\mu$ m) showing lower volume concentrations during the dry season and maxima in July. Spectral patterns of single scattering albedo (SSA) and complex refractive index also indicate coarse mode dominance conditions. However, FMF and fine mode volume fraction show values corresponding to fine mode dominance in terms of optical depth, and coarse mode dominance in terms of volume concentration during the dry season. In addition, AOD measurements below 0.15 are around 60% of the total, showing a cleaner atmosphere than sites located in the Sahel. AE dry season distribution is slightly bimodal (0.4 and 0.7 modal values) and symmetrical indicating a similar frequency for the two different particle populations (desert dust and background conditions). During the wet season AE shows two clear modes, a narrowed first mode centred in 0.15 (high dust events) and a smaller but wider mode centred in 0.4 (background conditions).

6

7 The aerosol-type characterization at Tamanrasset indicates desert mineral dust is the 8 prevailing aerosol. Large variations of AOD, AE almost inversely proportional to AOD and 9 Angstrom exponent difference ( $\delta AE$ ) negative or slightly positive have been observed 10 indicating higher extinctions linked to larger particles and dominance of one-particle mode. 11 Moreover, typical pure Saharan dust conditions have been observed, i.e. high-extinction 12 values (AOD>0.7) with AE<0.3 and  $\delta$ AE<0 corresponding to FMF<40% and fine mode radius (Rf) around 0.3µm. However, an anthropogenic fine mode contribution has been found 13 14 mixed with mineral dust (8.7% of total cases), i.e. AOD up to 0.4 for AE values ranging 15 between 0.6 and 1.1 and  $\delta AE$ , FMF and Rf between -0.3 and 0.2, 30% and 70%, and 0.10  $\mu m$ 16 and  $0.20 \,\mu\text{m}$ , respectively.

17

Potential sources of the air masses arriving at Tamanrasset have been identified: the triangle formed by Adrar des Ifoghas, Hoggar Mountains and Aïr Massif; a complex distribution of dust sources including northern Mali, northern Mauritania and the western flanks of Hoggar Mountains; and the eastern Libyan Desert. However, the Bodélé Depression has been found to be minor potential source at Tamanrasset. Dust uplift sources associated with summertime Mesoscale Convective System (MCS) events located south of Tamanrasset have been also identified.

25

Evidences of the arrival of fine particles to Tamanrasset during summer have been detected and nine events of polluted air masses coming from urban/industrial areas in Libya and Algeria have been shown.

29

#### 30 Acknowledgements

The AERONET sun photometer at Tamanrasset has been calibrated within AERONET-1 2 EUROPE TNA supported by PHOTONS and RIMA network and partially financed by the 3 European Community - Research Infrastructure Action under the Seventh Framework 4 Programme (FP7/2007-2013) "Capacities" specific programme for Integrating Activities, 5 ACTRIS Grant Agreement no. 262254. The authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion 6 7 model and READY website (http://ready.arl.noaa.gov) used in this publication. Financial 8 supports from the Spanish MINECO (projects of ref. CGL2011-23413, CGL2012-33576 and 9 CGL2012-37505) are also gratefully acknowledged. We are particularly grateful to the 10 Tamanrasset Global Atmospheric Watch (GAW) station's staff (l'Office National de la 11 Météorologie, Algeria) for supporting the measurement program. J.M. Baldasano and S. 12 Basart acknowledge the "Supercomputación and e-ciencia" Project (CSD2007-0050) from the 13 Consolider-Ingenio 2010 and Severo Ochoa (SEV-2011-00067) programs of the Spanish 14 Government. We also thank our colleague Dr. Celia Milford for proofreading the manuscript.

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1 Table 1. Mean K values (dimensionless) and standard deviation (Std. Dev.) for each channel

2	and for each period of correction <sup>a</sup> .

	1020 nm	870 nm	675 nm	440 nm	380 nm		
18 November 2007 to 22 March 2008							
K	0.9945	1.0085	1.0281	1.0716	1.1092		
Std. Dev.	0.0190	0.0200	0.0219	0.0257	0.0289		
23 March 2008 to 20 June 2008							
K	1.0674	1.0783	1.0943	1.1224	1.1600		
Std. Dev.	0.0093	0.0093	0.0079	0.0082	0.0090		

<sup>3</sup> <sup>a</sup>Data from 88 days fulfilling the requirements for applying the KCICLO method have been

4 used to compute the mean K values for the first period and 6 days for the second one.

Table 2. Least squares linear fit results (dimensionless) between NOOA hand-held sun photometer AOD measurements and three Cimel sun photometer AOD data sets (AERONET level 1.5 before and after KCICLO correction, and AERONET level 2.0). The parameters are the following: slope of the regression, Y intercept, correlation coefficient (R2), root-meansquare error (RMSE), and number of observations.

	Before KCICLO correction	After KCICLO       AERONET q         correction       assured (level)	
Slope	1.15±0.02	1.07±0.01	1.02±0.01
Y intercept	0.031±0.006	-0.014±0.004	$0.001 \pm 0.001$
$R^2$	0.968	0.981	0.983
RMSE	0.044	0.031	0.024
N. observations	450	450	1241

Table 3. Monthly means of aerosol optical depth (AOD), Angstrom exponent (AE). and
 precipitable water vapour (PWV) for the period October 2006 to February 2009 at

3 Tamanrasset<sup>a</sup>.

Month	AOD (440 nm) <sup>b</sup>	AE (440-670-870 nm) <sup>b</sup>	PWV (cm)	No. of days
January	0.07 (0.08)	0.69 (0.25)	0.37 (0.16)	93
February	0.12 (0.15)	0.49 (0.23)	0.48 (0.23)	66
March	0.23 (0.22)	0.31 (0.17)	0.57 (0.37)	62
April	0.40 (0.39)	0.19 (0.11)	0.64 (0.29)	60
May	0.37 (0.22)	0.15 (0.08)	0.99 (0.29)	62
June	0.43 (0.34)	0.17 (0.14)	0.97 (0.26)	60
July	0.39 (0.32)	0.32 (0.20)	1.15 (0.24)	62
August	0.41 (0.34)	0.44 (0.33)	1.39 (0.45)	62
September	0.33 (0.24)	0.36 (0.20)	1.22 (0.32)	61
October	0.20 (0.14)	0.41 (0.22)	1.01 (0.28)	93
November	0.10 (0.06)	0.54 (0.21)	0.68 (0.24)	90
December	0.09 (0.12)	0.72 (0.25)	0.49 (0.26)	93

- 4 <sup>a</sup>Corresponding standard deviations are shown in brackets.
- 5 <sup>b</sup>Dimensionless

1	Table 4. Statistics of aerosol	optical depth (AOD),	Angstrom exponent (	(AE). and precipitable
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	AOD (440) <sup>b</sup>	AE (440-670-870) <sup>b</sup>	PWV (cm)				
Dry season (342 days)							
Mean	0.09	0.62	0.51				
Std. Dev.	0.10	0.25	0.25				
Median	0.06	0.62	0.45				
Min.	0.01	0.08	0.06				
Max.	0.90	1.26	1.41				
Wet season (367 days)							
Mean	0.39	0.28	1.06				
Std. Dev.	0.31	0.22	0.40				
Median	0.29	0.20	1.03				
Min.	0.04	0.01	0.22				
Max.	2.18	1.28	2.71				

2 water vapour (PWV) from October 2006 to February 2009 at Tamanrasset<sup>a</sup>.

3 <sup>a</sup>Mean, standard deviation, median, minimum, maximum and number of days are shown for

4 the dry season (November-February) and for the wet season (April-September). March and

5 October are considered transition months.

6 <sup>b</sup>Dimensionless

- 1 Table 5. Monthly means of volume particle concentration (VolCon) of total, fine and coarse
- 2 mode, fine mode volume fraction  $(V_f/V_t)$ , and effective radius  $(R_{eff})$  for the period October

3 2006 to February 2009 at Tamanrasset<sup>a</sup>.

Month	VolCon (µm <sup>3</sup> µm <sup>-2</sup> )			Vf/Vt <sup>b</sup>	$R_{eff}$ (µm)			No. of days
	Total	Fine	Coarse		Total	Fine	Coarse	
January	0.04	0.005	0.04	0.21	0.63	0.163	1.92	38
January	(0.09)	(0.003)	(0.08)	(0.09)	(0.22)	(0.023)	(0.38)	38
February	0.06	0.008	0.05	0.17	0.70	0.151	1.89	27
i cordar y	(0.09)	(0.008)	(0.09)	(009)	(0.20)	(0.026)	(0.21)	21
March	0.17	0.015	0.15	0.11	0.78	0.141	1.86	22
Waren	(0.18)	(0.016)	(0.17)	(0.05)	(0.16)	(0.025)	(0.32)	
April	0.16	0.014	0.14	0.11	0.77	0.145	1.66	21
npm	(0.22)	(0.012)	(0.20)	(0.04)	(0.11)	(0.020)	(0.14)	21
May	0.23	0.017	0.21	0.09	0.86	0.133	1.72	24
ivituy	(0.18)	(0.011)	(0.17)	(0.02)	(0.11)	(0.012)	(0.09)	21
June	0.25	0.019	0.23	0.10	0.80	0.129	1.68	35
5 une	(0.22)	(0.008)	(0.22)	(0.04)	(0.19)	(0.016)	(0.13)	55
July	0.27	0.025	0.25	0.13	0.69	0.122	1.72	35
July	(0.31)	(0.014)	(0.30)	(0.06)	(0.22)	(0.014)	(0.11)	55
August	0.19	0.022	0.17	0.16	0.61	0.123	1.72	45
Tugust	(0.16)	(0.011)	(0.15)	(0.08)	(0.17)	(0.018)	(0.14)	15
September	0.20	0.018	0.18	0.10	0.79	0.139	1.62	23
September	(0.11)	(0.009)	(0.10)	(0.02)	(0.11)	(0.019)	(0.09)	25
October	0.12	0.014	0.11	0.13	0.71	0.143	1.62	45
October	(0.10)	(0.009)	(0.10)	(0.04)	(0.11)	(0.019)	(0.14)	15
November	0.05	0.008	0.04	0.19	0.58	0.146	1.77	54
	(0.03)	(0.005)	(0.03)	(0.07)	(0.13)	(0.024)	(0.27)	JT
December	0.04	0.007	0.03	0.22	0.59	0.159	1.82	38
December	(0.03)	(0.004)	(0.02)	(0.09)	(0.18)	(0.030)	(0.25)	50

4 <sup>a</sup>Corresponding standard deviations are shown in brackets.

5 <sup>b</sup>Dimensionless

- 1 Table 6. Seasonal means (dimensionless) of single scattering albedo (SSA), complex
- 2 refractive index (Real and Imag. Ref. Index), and Asymmetry parameter (Asym.) at 440, 675,

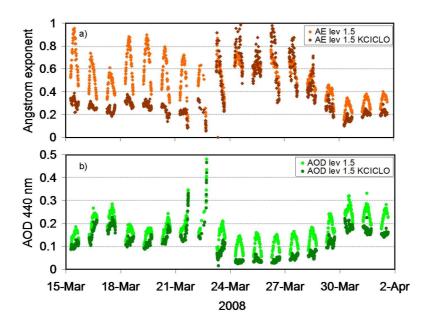
	Wet Season				Dry Season					
	440	675	870	1020	Ν	440	675	870	1020	N
SSA <sup>b</sup>	0.90	0.96	0.97	0.98	52	0.93	0.95	0.96	0.96	27
33A	(0.01)	(0.01)	(0.01)	(0.01)	53	(0.02)	(0.02)	(0.02)	(0.02)	
Real Ref.	1.45	1.47	1.44	1.43	52	1.41	1.42	1.42	1.42	27
Index <sup>b</sup>	(0.03)	(0.02)	(0.02)	(0.02)	53	(0.03)	(0.03)	(0.03)	(0.03)	
Imag. Ref. Index <sup>b</sup>	0.004	0.002	0.002	0.001	53	0.004	0.003	0.003	0.003	27
	(0.001)	(0.001)	(0.001)	(0.001)		(0.001)	(0.001)	(0.001)	(0.001)	
Asym. <sup>c</sup>	0.76	0.74	0.74	0.75	183	0.75	0.74	0.73	0.74	157
	(0.03)	(0.03)	(0.02)	(0.02)	103	(0.03)	(0.03)	(0.03)	(0.03)	137

3 870 and 1020 nm<sup>a</sup>. Number of daily available observations (N) is also indicated.

4 <sup>a</sup>Corresponding standard deviations are shown in brackets

5 <sup>b</sup>Level 2.0 for the wet season and level 1.5 filtered for the dry season

6 <sup>c</sup>Level 2.0 for the wet and the dry season



3

2 Figure 1. (a) Angstrom exponent (AE) in the range 440-870 nm and (b) aerosol optical depth

4 2008 (refer to legend for colour description). Two different corrections were applied before

(AOD) at 440 nm showed with and without KCICLO correction from 15 March to 1 April

5 and after 23 March. AE and AOD are dimensionless parameters.

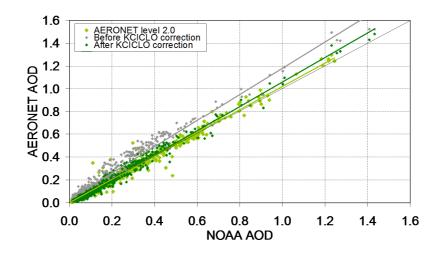
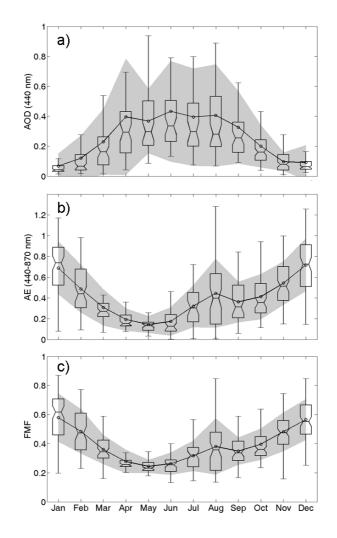


Figure 2. Dimensionless correlation between AERONET (Aerosol Robotic Network) aerosol optical depth (AOD) at 440 nm and NOAA (National Oceanic and Atmospheric Administration) AOD at 500 nm for time coincident data (within 15 minutes). The AERONET level 2.0 data (light green) cover the period from October 2006 to February 2009. The AERONET level 1.5 data from November 2007 to June 2008 are shown before (grey) and after (dark green) applying the KCICLO correction. Each data series is shown together with linear regression line. The solid black line is the 1:1 reference line.



1

Figure 3. Monthly box-and-whisker plot of daily (a) aerosol optical depth (AOD) at 440 nm, (b) Angstrom exponent (AE) in the range 440-870 nm, and (c) fine mode fraction (FMF) at 500 nm for the study period at Tamanrasset. Open dots are mean values; grey shaded area indicates the range of values between the mean plus or minus standard deviation; boxes show 25, median and 75 percentiles; and whiskers extend from each end of the box to the most extreme values within 1.5 times the interquartile range. AOD, AE and FMF are dimensionless parameters.

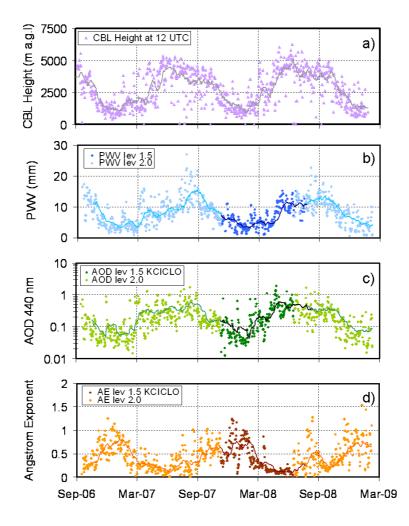
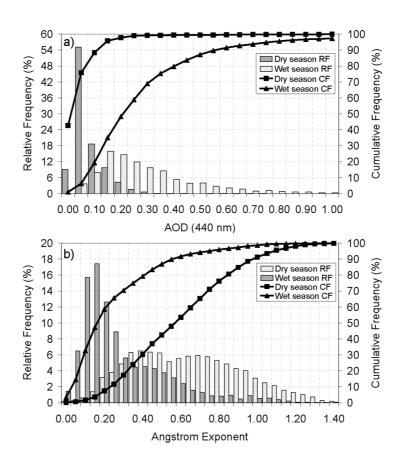
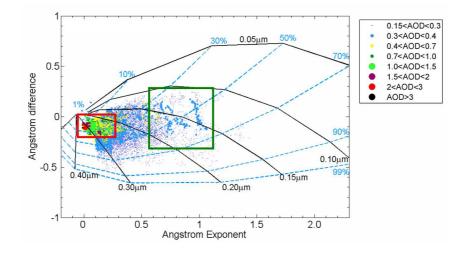


Figure 4. Time series of (a) Convective Boundary Layer (CBL) height [meters above ground level] determined from the 12 UTC soundings (violet triangles) in Tamanrasset (reprinted from Guirado et al., 2011), and AERONET (Aerosol Robotic Network) daily mean values of (b) precipitable water vapour (PWV) [mm], (c) aerosol optical depth (AOD) at 440 nm, and (d) Angstrom exponent (AE) in the range 440-870 nm (refer to legend for colour description). Solid lines correspond to 30-day moving averages. AOD and AE are dimensionless parameters.



2 Figure 5. Relative frequency (RF) and cumulative frequency (CF) of (a) aerosol optical depth

- 3 (AOD) at 440 nm and (b) Angstrom exponent (AE) in the range 440-870 nm at Tamanrasset.
- 4 Histograms are shown separately for the dry and the wet seasons (refer to legend for colour
- 5 and symbol description). AOD and AE are dimensionless parameters.



1

2 Figure 6. Angstrom exponent difference  $\delta AE = AE(440,675) - AE(675,870)$ , as a function of

3 Angstrom exponent (AE) and aerosol optical depth (AOD) (refer to legend for colour and

4 symbol description) at Tamanrasset site (10,460 observations) (reprinted from Guirado et al.,

5 2011). Strong dust events (red rectangle) and mixture of different aerosol types (green

6 rectangle) are indicated.  $\delta AE$ , AOD and AE are dimensionless parameters.

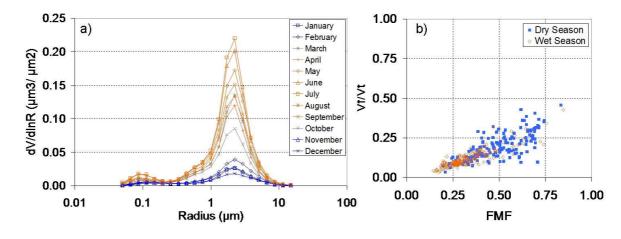
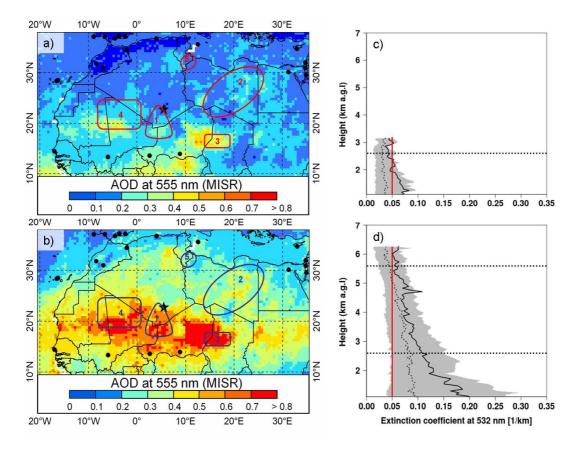


Figure 7. (a) Monthly means of aerosol particle size distribution  $[\mu m^3/\mu m^2]$  at Tamanrasset for the period 2006-2009. Same colours are used for the dry season (blue), the wet season (orange), and the transition months (grey). (b) Scatter plot of fine mode fraction (FMF) [dimensionless] and fine mode volume fraction (Vf/Vt) [dimensionless] for the dry and the wet seasons (157 and 183 coincident observations, respectively).





2 Figure 8. Averaged MISR (Multi-angle Imaging SpectroRadiometer) aerosol optical depth 3 (AOD) at 555 nm (dimensionless blue/red scale) for the period 2007-2008 during (a) the dry season (from November to February) and (b) the wet season (from April to September). 4 5 Geographical location of Tamanrasset (black star) in the Hoggar Mountains (Algeria) and present (2014) continuous monitoring AERONET (Aerosol Robotic Network) stations (black 6 7 dots) are indicated. Several potential dust sources, discussed in the text, have been identified 8 (solid red/blue lines) and numbered as follows: 1, triangle formed by Adrar des Ifoghas, 9 Hoggar and Aïr massifs; 2, eastern Libyan desert; 3, Bodélé Depression; 4, west Sahara region; and 5, Libyan-Tunisian border. Mean (black solid line) and median (black dashed 10 line) CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) extinction coefficients at 11 532 nm [km<sup>-1</sup>] are displayed for the period 2007-2008 during (c) the dry season (43 available 12 profiles) and (d) the wet season (95 available profiles) over Tamanrasset. Grey shaded area 13 14 shows the range of values between 20 and 80 percentiles. The red line marks the threshold of pristine conditions (extinction coefficient  $< 0.05 \text{ km}^{-1}$ ). Significant height levels, except the 15 16 ground level, for the Concentration Weighted Trajectory (CWT) analysis (2600 and 5600 m 17 a.g.l.) are marked (black dotted lines).

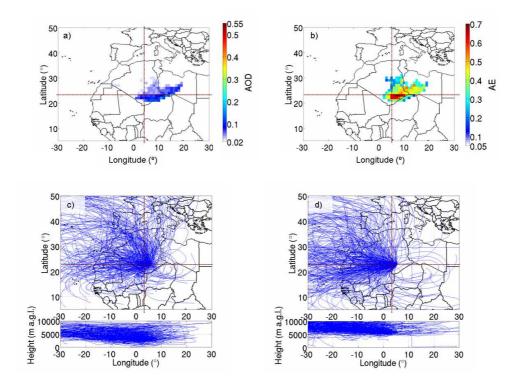


Figure 9. Concentration Weighted Trajectory (CWT) maps at ground level for (a) aerosol optical depth (AOD) and (b) Angstrom exponent (AE), and HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory Model) back-trajectories ending at (c) 2600 m a.g.l. and (d) 5600 m a.g.l. during the dry season (from November to February). Refer to dimensionless white/red scales for colour description of AOD and AE. Tamanrasset is located at the intersection of the four quadrants.

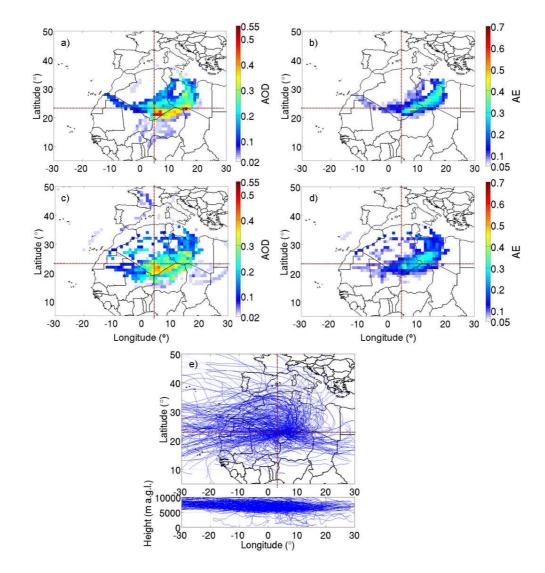


Figure 10. Concentration Weighted Trajectory (CWT) maps for aerosol optical depth (AOD) and Angstrom exponent (AE) at (a and b) ground level and (c and d) 2600 m a.g.l., and (e) HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory Model) back-trajectories ending at 5600 m a.g.l., during the wet season (from April to September). Refer to dimensionless white/red scales for colour description of AOD and AE. Tamanrasset is located at the intersection of the four quadrants.

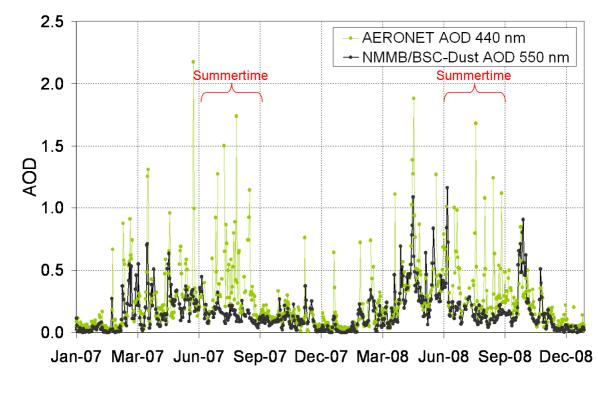
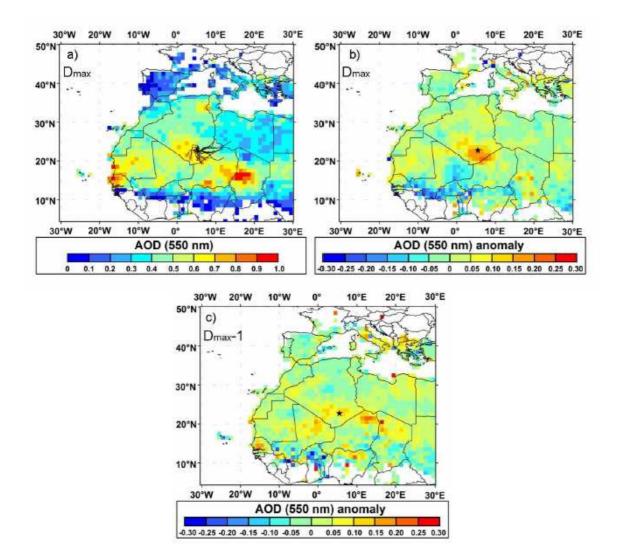


Figure 11. AERONET and NMMB/BSC-Dust AOD daily mean values for the period 2007-2008.



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Figure 12. Composite Moderate Resolution Imaging Spectrometer (MODIS) Deep Blue 550 nm (a) aerosol optical depth (AOD) and AOD averaged anomaly corresponding (b) to the 21 days of maximum ( $D_{max}$ ) AOD at Tamanrasset during Mesoscale Convective System (MCS) events, and (c) to the corresponding previous days ( $D_{max}$ -1). Tamanrasset station is marked with a black star. Two-day HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory Model) back-trajectories arriving at Tamanrasset at ground level (black solid lines) are also displayed in panel (a).

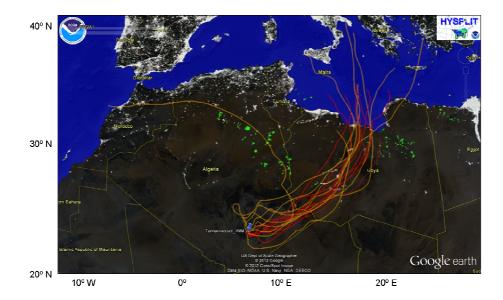


Figure 13. HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory Model) backtrajectories arriving at Tamanrasset (blue pin) at ground level (red lines) and 2600 m a.g.l.
(yellow lines) are displayed for several case studies. Defense Meteorological Satellite
Program (DMSP) Nighttime Lights (shown as background) identify gas flares by green
colour.