

## Year-round study of the optical properties of airborne dust in Antarctica

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**Summary.** — I report experimental results obtained from OPTAIR, a multidisciplinary project to study the optical properties of airborne particles at Concordia Station, on the East Antarctic plateau, with the aim to assess the relationship among the optical properties of dust suspended in air and deposited on ground by the snow. A permanent instrument based on the Single-Particle Extinction and Scattering (SPES) method specifically designed and realized has been installed in November 2018 and continuously produces time-resolved data, providing several optical properties for each particle. The aim is to feed the models describing radiation transfer through the Earth's atmosphere, an open issue for what concerns the effects of dust. Data show evidence of important changes of the optical properties of dust across the year, with a relevant fraction of particles accumulated during short bursts lasting a few hours. This shows the need of time resolved information about the optical properties of dust to infer the effective impact of dust on radiative transfer.

### 1. – Introduction

Understanding the present-day optical properties of dust, the processes related to transport and the temporal evolution of dust load are essential elements for a deeper understanding of past climate conditions through mineral dust aerosol analyses in firn and ice cores. The successful drilling program EPICA-Dome C (EDC) provided the oldest ice core with an archive of climate and atmospheric conditions over the last 800000 years from the central East Antarctic plateau (Dome C). The analysis of the EDC ice core clearly showed that dust deposited in the past on the East Antarctic plateau derives from South America and that dust concentration in ice (and in the DomeC atmosphere) is influenced by climate conditions at the sources. Because of natural climate changes and the seasonality of dust emissions [1-3] the dust flux remarkably changed, dropping

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down to extremely low levels many times as in the Holocene and modern times. Average depositional fluxes of mineral dust are the lowest on Earth, about  $0.2\text{--}0.4\text{ mg m}^{-2}\text{ yr}^{-1}$ . Moreover, the dust reaching DomeC is almost free from any anthropic pollution, making this site ideal and unique to better understand the relationship among the properties of dust suspended in air and deposited on ground.

Besides the chemical and physico-chemical properties, that are of utmost importance to characterize the sources and the evolution of dust travelling through the atmosphere, the optical properties are those ultimately responsible for the radiative effects on sunlight. Radiative transfer is one of the key elements determining climate, widely recognized to be affected by aerosols in general [4]. Unfortunately, optical properties of dust and aerosol are not well understood nor characterized, also because of their strong heterogeneous nature and the size range that is very critical to be described quantitatively precisely enough. Mineral dust, marine aerosol, volcanic products and products of anthropic pollution are mixed up in a very complex system undergoing strong temporal evolution. Fixing parameters that allow models to be precise enough for describing the climate is currently impossible due to the lack of knowledge of optical properties and their influence on radiative transfer. Moreover, the dust from the ice core archives is typically analyzed when suspended in meltwater, thus causing the loss of soluble elements and unavoidably altering the optical properties of each measured particle with respect to those occurring at the time when it was suspended in the atmosphere of the past. Finally, information is missing also with respect to the processes of deposition, that drives the dust from the atmosphere to the ground and fix it into firn and ice for millennia. Elucidating the relationship among airborne dust in terms of the optical properties is still a challenging issue.

Because of the very low dust load in the Antarctic atmosphere and the extremely low concentration of dust in ice cores in the modern times, the quantitative assessment of dust optical properties is very challenging. This limitation essentially prevents from the use of methods accessing the information coming from a collection of many particles, thus limiting dramatically the number of available techniques. Nevertheless, an advantage emerges from this strong limit, since interpreting data from a collection of particles always requires the use of ill-posed problems and hides some other issues related to the interpretation models adopted. Among the others, particle size is an important factor affecting dust radiative properties and it is usually the one parameter available. To date, accurate and precise measurements of (equivalent spherical) dust size distribution and concentration can be obtained through the well-established Coulter Counter method, although single-particle obscuration sensor (SPOS) method is often used in continuous flow analyses. Results from these two methods often disagree, probably because of the different parameters accessed [5]. In any case, other features affect light scattering as composition, internal structure, shape, surface roughness etc., making the size parameter not enough to provide the necessary information for the radiative transfer models. Some of this information can be obtained through microscopy (optical, scanning and transmission electron microscopy) and geochemical analyses. A consistent working time is required in sample preparation, measuring and data analysis, with implications for the validity of statistics that can be derived from a very limited number of samples. Moreover, the same dependence of particle radiative properties upon many parameters introduces a further limitation: even with extreme measurement accuracy of single particles, recovering the size from any measured parameter needs a model relating each signal to one particle size. This process, known as “inversion”, dramatically depends upon the validity of the model, until the same concept of “size” is questionable since it can be only

associated to a spherical equivalent of the particle [6]. Dust grains are not spherical, so that the size is not defined and will be recovered in a way that unavoidably contains some errors. An example of the possible errors in this inversion process has been recently reported just for Antarctic dust [7].

From all the above it emerges that addressing the issue of radiative properties of dust can be attempted by following a very tight pathway: i) measuring the optical properties needed to the radiative transfer models in the most direct way, meaning not including inversion processes, that are unavoidably model dependent; ii) performing precise measurements on single particles, obtaining the properties of the collection from those of each grain; iii) measuring more parameters from the same particle at a time, thus allowing a multiparametric analysis with tight constraints when interpreting data.

Stemming from these needs I designed a specific project, based on an innovative optical method to measure the optical properties of single particles. It aims to perform a long-term monitoring of dust at DomeC, collected both from the air, in real time, and from the ground on a regularly time base. In the following sections I briefly introduce the instrument and the preliminary results obtained for the optical properties throughout the year 2019.

## 2. – The project OPTAIR: methods

OPTAIR is a multidisciplinary project aimed at determining and studying the optical properties of airborne particles at Concordia Station (DomeC, East Antarctic Plateau). The project has been funded by the Italian National Antarctic Program (PNRA). A permanent instrumentation has been installed in fall 2018 and is currently working continuously, measuring single airborne particles overall the year. In parallel, the deposited snow has been collected from the ground with a regular frequency starting from the beginning of year 2021 (not before due to logistic issues). The snow will be accurately characterized in European laboratories and put in correlation with the data from airborne particles and with data from continuous LIDAR measurements, performed at Concordia since years. The goal of the project OPTAIR is to obtain on an experimental basis the optical properties of airborne and deposited dust and to assess the impact on past and present climate.

OPTAIR is centered on the use of the novel Single-Particle Extinction and Scattering (SPES) optical method [8]. The advantage of the method is represented by the capability of measuring both the extinction cross section,  $C_{\text{ext}}$ , and the forward scattering fluence,  $F(0)$  of each particle sensed by the instrument. The optical layout relies on self-reference interference of the scattered and the transmitted light, making it extremely robust and rigorously calibration free. Moreover, two additional sensors collect light at  $45^\circ$  and  $90^\circ$ , thus measuring the corresponding fluences,  $F(45)$  and  $F(90)$ . In fig. 1 a schematic of the optical layout is depicted. A collimated laser beam (wavelength  $\lambda = 640$  nm, power 40 mW) is expanded 2.5 times by a Galilean telescope composed by two achromats A1, A2 (focal lengths  $f_1 = -20$  mm,  $f_2 = 50$  mm) and the beam is focused by the doublet A3 (focal length  $f_3 = 50$  mm) into a flow cell driving the air from outside through an inlet. The focal spot is characterized by a beam waist  $w_0 = 15$   $\mu\text{m}$ . The air is driven through a thin sheet, 100  $\mu\text{m}$  thick, that allows the particles to be illuminated under well-known conditions. The forward scattered light passes through the attenuator A (OD1), then collected at zero angle, where it is superimposed and interferes with the transmitted beam onto a quadrant photodiode (First Sensor QP50). A specific front-end electronics (FE) handles the photocurrents before being converting into voltages. The

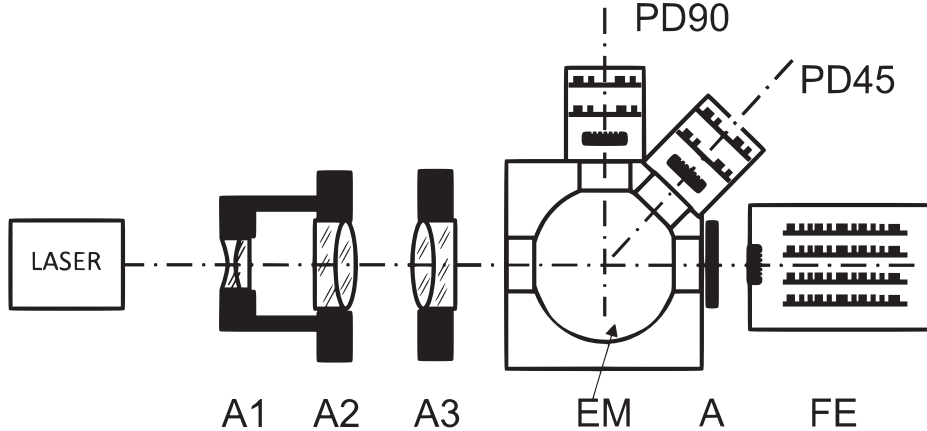


Fig. 1. – Schematic layout of the instrument installed at DomeC. A1, A2, A3 are lens doublets; EM: elliptical mirror; A: OD1 attenuator; FE: front-end electronics; PD45, PD90: photodiodes. See text for details.

beam intensity is monitored slowly (100  $\mu$ s characteristic constant) with respect to the transit times (a few  $\mu$ s each). A current supplier provides an opposite current to sum with the measured photocurrent, so to bring the total to vanishing values: the fast fluctuations due to the particles passing through the beam and the current generated by the supplier are converted into voltages and sent to separate digitizers. Therefore, the fast fluctuating signals, down to  $10^{-4}$  of the total intensity, can be sent to a fast 12 bit digitizer that exploits the whole dynamics to provide the raw data from the four photodiodes. The slow signals provide a continuous monitoring of the laser beam, thus allowing the system to be always automatically calibrated. Scattered light is also collected at  $45^\circ$  within a solid angle of 0.1 sr by an extended photodiode (PD45, 8mm in diameter) and at  $90^\circ$  by an elliptical mirror EM covering a solid angle of 1.2 sr. The foci of the ellipsoid are such that the focal spot of the laser, that is the point where the light is scattered, is conjugated with a small aperture (1 mm in diameter) in front of the photodiode PD90. Thus a confocal geometry prevents most of the stray light to affect the measurement. The device and the corresponding data analysis have been extensively described in [9].

Each particle is associated with four independent physical quantities. From the forward-scattering measurements we can obtain the dimensionless complex scattering amplitude,  $S(0) = \text{Re}S(0) + i\text{Im}S(0)$ , defined in such a way that  $F(0) = |S(0)|^2$  and  $C_{\text{ext}} = 4\pi/k^2\text{Re}S(0)$ . They can be converted into some physical quantities of importance to characterize the radiative properties of the particle in a way that is almost independent of the particle size. The *optical thickness* of the particle is defined as  $\rho = 2\text{Re}S(0)/\text{Im}S(0)$  and two parameters that are the unnormalized phase factors at  $45^\circ$  and  $90^\circ$  are defined as  $P(\theta) = F(\theta)/k^2C_{\text{ext}}$ , where  $\theta = 45^\circ, 90^\circ$  is the angle. Of course, the quantities  $F(0)$  and  $\rho$  are not independent, the number of independent parameters being four. One of the most striking advantages of this method relies on the measure of  $F(0)$  itself, an almost unique opportunity that poses an important constraint to determining the phase factors. I stress that the normalized phase factor  $p(\theta)$  cannot be obtained, just because the device does not provide the measurement of the single scattering albedo of the particle,  $ssa$ , so that  $P(\theta) = ssa p(\theta)$ . Nevertheless, for the purposes of characterizing the scattering features, the value  $ssa$  does not need to be fixed with

extreme precision, since it changes around 0.8–0.95 typically, introducing an error of less than 15% in evaluating  $p(\theta)$  from  $P(\theta)$  by simply imposing a reasonable value. This is not the case, of course, if one needs to estimate the absorption properties: in this case the same range will give a 300% error.

As mentioned above, I aim to recover the optical properties with null or minimum model dependence and the above procedure permits to do so. Nevertheless, for the sake of comparison with other approaches, the particle size distribution can be obtained by inverting the extinction data, as usually done. In comparison to obtaining size from  $90^\circ$  scattering data, the size obtained from  $C_{\text{ext}}$  is the most independent of shape or any other nonideality of the spherical interpretation. As an example, by comparing the sizes obtained for a given particle population from  $C_{\text{ext}}$  and from  $F(90)$ , the latter being what is usually done in the Optical Particle Counters, the two size distributions show remarkable discrepancies. Comparing the two radii gives an important figure of merit to quantify the deviation from the ideal spherical model.

When plotting data on a log-log scale, all parameters exhibit nice normal distributions. Data have then been analyzed through statistical estimators properly introduced for lognormal stochastic distributions. Let us say  $X$  is the variable to be studied, with a lognormal distribution given by  $X = e^{\mu + \sigma Z}$ , where  $Z$  is a stochastic variable with normal distribution with mean  $\mu$  and standard deviation  $\sigma$ . Data can easily be described by the geometric median  $M = e^\mu$  and the geometric standard deviation  $G = e^\sigma$ , that allow to properly quantify the changes in the different data sets considered below. The immediate meaning of these estimators is, for example, that 67% of the counts are within the interval  $(M/D - M \cdot D)$ .

The SPES method can also be easily operated on liquid suspensions. In recent years it demonstrated to be effective in determining properties of particles such as shape [10] and internal structure [11, 12] in the case of liquid suspensions. Exploited on the so-called “old” DomeC ice core, it brought clear evidence of the shape variability of dust with time, with important impact on the radiative transfer of the atmosphere in the past [13]. Operating the SPES method in liquid to analyze the dust content of snow accumulated simultaneously with the airborne particle measurements at the same site will represent a unique feature of this work.

The instrument has been installed within the Atmos shelter at Concordia Station, DomeC. A dedicated inlet brings the air from outside into the instrument thanks to a pump, with a flux of approximately 5 liters per hour. The air passes through a warming pipe that increases the air temperature of tens of degrees, thus sublimating the ice coverage that can be there around the dust grains. It is completely automated, controlled and reconfigurable from remote: this constitutes a very convenient situation for a device operating in a remote region and almost unattended.

Two procedures are adopted to select the data to be analyzed: 1) the data collected when the wind was coming from a direction that could in some way bring pollution from the station power plant have been removed, and the corresponding data analyzed separately for the sake of comparison; 2) thanks to the intrinsic validation of the SPES method (see [8]), the particles passing through the focal region within 1/10 of the beam diameter have been selected, so to guarantee a rigorous model-independent set of raw signals to produce the data.

In the following section I will briefly sketch the main results obtained by a preliminary data analysis performed on data collected during the entire 2019.

TABLE I. – Numerical values of the optical properties during the year 2019. The first column indicates the season, with the capital letter of each month. October is missing. The last two lines contain the results obtained by separating the spike events recognized by the LIDAR, group S, from the rest of the sample, N.

Season	$\langle C_{ext} \rangle \mu\text{m}^2$	$\langle a \rangle \mu\text{m}$	$P(0)$	$P(45)$	$P(90)$	$\rho$
JF	2.95	0.51	8.36	0.121	0.0165	1.35
MAM	2.30	0.45	7.71	0.101	0.0136	1.15
JJA	1.78	0.41	5.74	0.131	0.0203	1.01
SN	3.36	0.52	12.86	0.147	0.0186	1.25
S	1.75	0.41	5.35	0.132	0.0214	1.01
N	2.43	0.46	8.06	0.120	0.0159	1.15

### 3. – Results

Two features emerge clearly by first analyzing the number counts per day: a small variability is observed across the year, with a maximum in winter; moreover, data exhibit sudden increases lasting hours, a couple of days maximum, when counts increase a factor 10 or larger (spikes). By removing these events the “background” count rate is more uniform across the year. Remarkably, the amount of particles accumulated during these events is more than 1/3 of the total, despite the short time integration, less than 10%. This result clearly shows the need for a high-resolution characterization of the dust accumulation to estimate the actual effect of the dust load in atmosphere. I will first report the results obtained from the seasonal analysis, later on I will report a preliminary discussion of the spike events.

**3.1. Seasonal analysis.** – By considering the optical properties discussed above, the following can be assessed. The extinction cross section  $C_{ext}$  shows a minimum during the winter, down to  $-55\%$ . This corresponds to a size reduction of about 20%, although the figure of merit for the validity of the spherical approximation,  $r$  changes appreciably indicating that another feature besides the size, likely the shape, is changing. The optical thickness  $\rho$  decreases as well, down to  $-67\%$  during the winter. On the contrary, all the unnormalized phase factors  $P(0)$ ,  $P(45)$ ,  $P(90)$  show almost no changes, the only variability being a relatively small  $+28\%$  for  $P(0)$  during the winter. The numerical values of the optical properties are reported in table I.

The geometric standard deviations of all quantities show very small changes, except for the winter (JJA) when the distributions appear slightly shrunk and for parameter  $\rho$ , that shows a minimum spread during the same period.

**3.2. Spike events.** – The analysis of LIDAR data, more precisely the signals from depolarized backscattered light that indicates the presence of dust, show a very interesting and marked correlation with spikes: they occur together with dust plumes descending from an altitude of 1–2 km to the ground in some hours. An analysis of the meteorological explanation of this effect is beyond the scope of this work. Here I limit to select OPTAIR data collected during those days with subsidence detected from the LIDAR, so to adopt a completely blind procedure for the data choice. I will group all particles collected during subsidence events, indicated here below as “S”, and those occurring dur-

TABLE II. – Results obtained by fitting the function (2) to data reported in table I, calculated following eqs. (3), (4), (5).

Season	$g_1$	$g_2$	$f$	$Pb$	$g$
JF	0.90	0.60	0.959	0.041	0.819
MAM	0.89	0.66	0.967	0.033	0.834
JJA	0.88	0.54	0.945	0.055	0.778
SN	0.92	0.59	0.960	0.040	0.824
S	0.88	0.45	0.947	0.053	0.783
N	0.90	0.60	0.959	0.041	0.819

ing the complementary time (excluding the wind from the station, as explained above), “N” group. Quantitatively speaking, S group contains more than 30% of the particles collected during less than 10% of time. The optical properties (and average size) of particles detected during the S events are remarkably different from the complementary group N, an important signature in view of refining radiative transfer models. Results are reported in the last two lines of table II.

Data show a remarkable difference between the two conditions, probably ascribable to the different apportionment driving the dust particles to DomeC. The extinction cross section differs by 40%, and by comparing  $P(0)$  and  $P(90)$  in the two cases it is evident how the scattering functions change, being more forward directed in case N than in case S. In principle, this could simply be ascribed to a change of size, smaller in the S case than N. Nevertheless, by comparing the sizes obtained from  $C_{\text{ext}}$  to those obtained from  $P(90)$ , an additional difference appears to be needed, the size only being unable to explain this discrepancy. This confirms the inability of one parameter to properly give the size, that underlines the importance of working without inverting to size the optical data. Finally, comparing the results obtained from the two analyses performed here, a noticeable similarity between the S case and the results obtained during winter, JJA. This is consistent with the occurrence of subsidence events mainly in winter.

#### 4. – Data analysis

Here I sketch how to recover some information of interest for the radiative transfer models. Besides the absolute optical depth, that is easily measured by other methods and in our case can be directly related to the rigorously measured  $C_{\text{ext}}$  and the number density of the scatterers, here I focus on the asymmetry parameter that is much usually more difficult to determine for single particles. As introduced in [9], the information about the phase factor, even if unnormalized, allows to give an estimate about the asymmetry parameter for a collection of particles. This factor is usually defined as the average cosine of the scattering angle  $\theta$  weighted for the phase function, that is

$$(1) \quad g = 2\pi \int_0^\pi p(\theta) \cos \theta d\theta.$$

It is worth noticing the formal equivalence of the asymmetry parameter  $g$ , properly averaged over a given population of scatterers, and the ratio between the net flux and the average intensity, quantities defined for example as in Chandrasekhar [14]. Hence the



importance to assess the asymmetry parameter for a collection of polydisperse particles in view of radiative transfer models, a task that becomes feasible with OPTAIR data.

In order to assess this parameter from our experimental data, in [9] we introduced a modified phase function with respect to the usually adopted Henyey-Greenstein (HG) approximation, which definitely does not fit the experimental data. In this previous work an exponential tail was added to the common HG function. Here, by following the works by Reynolds [15] and Sorensen [16], for example, I introduce the modified phase function obtained as the sum of two Gegenbauer functions (that include the HG as a particular case). I use the sum of a traditional HG function and an additional term chosen to reproduce the well-known Porod law for the decrease of the scattered intensity as a function of the angle [16]. I will indicate this function as HGP for brevity. The two terms are equally weighted and the phase function reads

$$(2) \quad p(\theta) = \frac{1}{2} \frac{1 - g_1^2}{(1 + g_1^2 - 2g_1 \cos \theta)^{3/2}} + \frac{1}{2} \frac{(1 - g_2^2)^2}{(1 + g_2^2 - 2g_2 \cos \theta)^2}.$$

Here,  $g_1$  and  $g_2$  act as the only two fit parameters. Actually, also the relative weight could be fitted to data, but we keep it constant to 1/2 each. This choice follows from the considerations discussed in [16], showing that actually the phase function of each particle does contain both contributions, hence they do not come from different fractions of the population. Function (2) is written in such a way to be already integrated over the azimuthal angle. It can now be easily integrated analytically to give the forward and backward scattering,  $f$  and  $b$  defined below, and the asymmetry parameter  $g$ , all quantities depending upon the fit parameters  $g_1$  and  $g_2$ . Therefore

$$(3) \quad f = \int_0^{\pi/2} p(\theta) d \cos \theta = \frac{(g_1 + 1)(\sqrt{1 + g_1^2} + g_1 - 1)}{4g_1 \sqrt{1 + g_1^2}} + \frac{(1 + g_2^2)^2}{4(1 + g_2^2)^2},$$

$$(4) \quad b = \int_{\pi/2}^{\pi} p(\theta) d \cos \theta = \frac{(g_1 - 1)(\sqrt{1 + g_1^2} - g_1 - 1)}{4g_1 \sqrt{1 + g_1^2}} + \frac{(1 - g_2^2)^2}{4(1 + g_2^2)^2},$$

$$(5) \quad g = \frac{1}{2} \frac{2g_2(1 + g_1^2) + (1 - g_2^2)^2 \log(1 - g_2^2) - (1 - g_2^2)^2 \log(1 + g_2)}{4g_2^2}.$$

By fitting to data in table I, we find the results reported in table II.

Again, these optical properties depend upon time and also change from S to N group. In particular, the backscattered fraction increases up to 30% from N to S, accordingly to the decreasing  $P(0)$  mentioned above. The importance of the measured changes in the optical properties can be appreciated by considering the product of the extinction cross section  $C_{\text{ext}}$  and the  $f$  and  $b$  fractions. If one considers  $C_{\text{ext}}$  and  $C_{\text{sca}}$  as equal, an approximation that can be done within the assumption that  $ssa = 1$  that is reasonably within 5%, the product gives the efficiency of the ‘‘average scatterer’’ to generate forward and backward scattering. Therefore, one can notice that while  $f$  is very close to unity and its changes are relatively small (3%), the contrary is true for  $b$  that changes about 60%. On the contrary, the cross section changes in the opposite direction, but not enough to compensate the change in the backscattering efficiency. This means that the optical properties influence appreciably the diffuse reflection of the dust in the atmosphere. Again, the effect of size only is not enough to explain this behaviour, that is exquisitely ascribable to the specific features of dust, making it deviating from the ideal spherical approximation.



As a final remark, I point out that for dust collected during Antarctic winter, group JJA, is actually to be considered that no Sun is there, so that i) the huge amount of particles and ii) the peculiar optical properties must be considered to be ineffective in view of any radiative transfer effect. This poses important caveat for studies performed on dust collected from ice cores, especially from the elder times, where time resolution is not enough to associate some specific dust properties to the effective solar illumination, therefore to properly describe the true effect of dust. Especially during glacial periods, when dust load is much higher, this can be a limitation to be carefully considered.

## 5. – Conclusions

I have briefly reported about the preliminary result obtained by analyzing data collected during the year 2019, a set of data that will be enriched by further data and observation thanks to the continuous monitoring at DomeC within the project OPTAIR. Data clearly show the need for a multiparametric, single-particle approach to overcome the current difficulties of interpreting data and/or predicting radiative transfer properties of dust. As for aerosol in general, the size range of airborne dust affecting sunlight is just around the wavelength, in the so-called intermediate size range, where the distributions of the internal fields within the scatterers are particularly difficult to be described and influence the radiated fields appreciably, raising the issue that modelling radiative properties of the particles is extremely difficult. As a result, from one side the accuracy in sizing the dust is appreciably limited even with instrumentation working on single particles, on the other side it drastically prevents the models to be predictive of the real effects caused by dust on sunlight. Hence the tentative approach to proceed without inverting experimental data to sizes and to overcome this issue by measuring the optical properties directly. The tentative approach introduced here to recover the asymmetry factors can be largely ameliorated on the basis of more accurate analysis of the typical phase functions for dust.

Due to the number of features that affect the optical properties of dust, including size, shape, orientation, composition, internal structure, optical methods are the only solution for trying to assess the true effects of light on airborne dust or aerosols in the real cases. As a matter of fact this information is needed, for example to address the recent directives about climate change. The so-called “Climate actions”, for example from the 13th Goal of the ONU 2030 Agenda, aims to develop two concepts to reduce and possibly prevent the consequences of climate change: *mitigation and adaptation*. Ultimately, to properly act in this direction one of the most important ingredient will be to better understand the Earth’s energy balance and to extend environmental monitoring solutions to provide strong based data. Although an innovative generation of Earth monitoring satellites is already operating an accurate remote sensing from space, data collection needs to be improved on ground, or from ground through remote sensing. Both approaches definitely need to better understand light scattering and radiative properties of small airborne particles.

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