Mem. S.A.It. Suppl. Vol. 16, 119

Memorie della
Supplementi

DUSTER (Dust in the Upper Stratosphere Tracking Experiment and Retrieval)

PRELIMINARY ANALYSIS

A. Ciucci[1,2], P. Palumbo[1], R. Brunetto[1,3], V. Della Corte[1,5], S. De Angelis[1,2],
A. Rotundi[1], F.J.M. Rietmeijer[4], E. Zona[1], L. Colangeli[9], F. Esposito[5],
E. Mazzotta Epifani[5], V. Mennella[5], S. Inarta[5], S. Peterzen[6], S. Masi[7], and R. Ibba[8]

[1] Dip Scienze Applicate, Università di Napoli 'Parthenope', Centro Direzionale 80143 Napoli, Italy, email: alessandra.ciucci@uniparthenope.it
[2] Dipartimento Ingegneria Aerospaziale, Università di Napoli Federico II 80143 Napoli, Italy
[3] European Space Astronomy Centre, 28691 Villanueva de la Cañada, Madrid, Spain
[4] Department of Earth and Planetary Sciences, University of New Mexico, Albuquerque, NM 87131-0001, USA
[5] INAF - Osservatorio Astronomico di Capodimonte 80131 Napoli, Italy
[6] ISTAR (International Science Technology And Research) Pagosa Springs, Colorado USA
[7] Dipartimento di Fisica, Universitá La Sapienza, 00100 Roma, Italy
[8] ASI (Agenzia Spaziale Italiana)
[9] Solar System Division, Research and Scientific Support Department, European Space Agency, ESTEC, Keplerlaan 1, PO Box 299, 2200 AG Noordwijk ZH, The Netherlands

Abstract. The DUSTER project is aimed at uncontaminated collection and retrieval of stratospheric solid aerosol particles, in the submicron/micron range. The Earth stratosphere contains extraterrestrial dust, dust from natural and anthropogenic activities. Our main target is the study of dust originated in our planetary system. We present here the preliminary results of the June 2008 campaign. After recovery, collected particles were identified by comparing FESEM images taken on the pre-flight collector with those obtained on the post-fight collector. Possible contamination was monitored by FESEM observation of the 'Blank'. Morphology, dimension, and composition of collected particles were defined using a FESEM equipped with an EDX system. The collected sample are in the size range 0.5-150 μm, $\approx$ 30% of aerosols sizing $0.5 - 1.5$ μm, a range poorly studied so far.

Key words. Stratospheric dust – laboratory analyses – FESEM-EDX

1. Introduction

At any given time stratospheric dust includes: natural terrestrial dust (e.g. volcanic dust,

---

Send offprint requests to: A. Ciucci

windblown dust, biomass burning), dust from anthropogenic activities (e.g. coal and fuel oil burning by aircraft), and extraterrestrial dust. The study of stratospheric natural and anthropogenic dust is an important tool to better understand atmospheric movements and the impact on climate, the connection between atmospheric dust layers and the potentially increasing levels of contamination by anthropogenic activities.

The extraterrestrial dust component may include interplanetary dust particles from asteroids and comets, and possibly interstellar dust (Flynn 1997). Interplanetary dust in the Zodiacal cloud is permanently replenished by dust mostly ejected from cometary nuclei, and from collisions in the asteroid and Kuiper belt (Divine 1993; Grun et al. 2001).

Stratospheric dust and aerosol can be studied by remote sensing (e.g. LIDAR, Deshler et al. 2003), in-situ (e.g. OPC, Kasai et al. 2003), or sample collection and analyses. The stratospheric aerosol layer was for the first time measured in late 1950's (Junge et al. 1961) by in-situ measurements and sampling performed with instruments carried aloft on stratospheric balloons up to 30 *km*. Several particle collections have been carried out in the stratosphere in the past 40 years. NASA has carried out a long-term program of collection in the lower stratosphere ($\approx$ 20 *km*) by aircraft flights, with the main goal of collecting cosmic dust (Warren & Zolensky 1994), using flat-plate collectors coated by a high viscosity silicone oil layer. The collected interplanetary dust particles have typical sizes of tens of micrometers. They are morphologically, chemically and mineralogically distinct from any type of natural or anthropogenic dust (Rietmeijer 1998, 2002).

The efforts to collect and characterize dust collected from the stratosphere all involved several steps in laboratory handling to remove the collected dust and prepare them for laboratory analyses, which heightened the risk of contamination and potentially damage fragile particles when removing silicone oil. Inertial-impact collection of stratospheric dust by direct deposition onto substrates that fitted in sample holders of scanning and transmission electron microscopes showed that collected dust could be successfully characterized (Testa et al. 1990; Rietmeijer 1993). The experience gained from these dust collection efforts showed a need for a collector that minimized extraneous contamination and that could routinely sample the stratospheric dust population. To achieve these goals our group recently developed a new instrument called DUSTER (Dust in the Upper Stratosphere Tracking Experiment and Retrieval), aimed at uncontaminated collection and retrieval of stratospheric solid aerosol particles (called dust), in the submicron to micron range (Palumbo et al. 2008). Collections are carried out at about 40 *km*, using stratospheric balloon flight opportunities to carry DUSTER aloft.

DUSTER is specifically designed to minimize contamination, by reducing sample manipulation. It will provide the following critical information on solid aerosol: 1) relative abundances of different components; 2) efficiency of stratosphere injection from various dust-producing sources, residence times and stratosphere mixing; 3) their chemical and physical properties; 4) direct (interaction with radiation, chemical and physical stability) and indirect (aerosol as catalyst of chemical reaction and influence on local physical and chemical conditions) effect on atmospheric chemistry and radiation budget.

In this paper we report the first successful collection of stratospheric dust at $\approx$ 37 *km* by DUSTER. Preliminary analyses of the grains is performed in the LFCP Laboratory (Naples, Italy) using Field Emission Scanning Electron Microscope (FESEM) equipped with Energy Dispersive X-Ray analysis (EDX).

2. DUSTER instrument

The prototype of DUSTER was constructed in 2006. It was a box of $(0.6 \times 0.6 \times 0.46)$ m^3 weighting 65 *kg*. This instrument was launched in January 2006 during a CNES (French Space Agency) balloon flight campaign from Esrange, Kiruna (Sweden). During this test flight, DUSTER collected grains for 2 *h* at a floating altitude of $\approx$ 27 *km*.

A modified DUSTER (DUSTER2008) instrument was launched in June 2008 thanks to a dedicated campaign by ASI (Italian Space Agency) from Longyearbyen, Svalbard (Norway). DUSTER2008 had a significantly reduced size compared to its prototype: it sized $(0.4 \times 0.4 \times 0.3)\ m^3$ for a weight of 30 *kg*. The instrument remained in the stratosphere for 3.5 days and it was switched on for 55 *h* collecting particles at $\approx$ 37 *km*, before being recovered in Thule (Greenland). Details on instrument design and requirements are discussed by (Della Corte et al. 2011).

The actual collector and the blank control sample are two identical sample holders: the former to collect stratospheric particles, the latter to monitor the environment, before flight, during stratospheric collection, and at every relocation of the two.

The sample holder was designed with a smooth surface that is easy to keep clean, appropriate to analyse the collected dust particles with different instruments but without manipulation to minimize contamination. It consists of a round stub whose gold smooth surface is pierced by fourteen holes proper to accommodate fourteen grids for TEM (Transmission Electron Microscope) and FESEM analysis. The system is fits together by fourteen pins and a base in stainless steel. Particles are collected by direct deposition on the gold TEM grids supporting a holey carbon film onto which the dust is directly deposited.

These TEM grids are suitable for microscopic identification, as their mesh can be numbered with respect to a reference position. Before the assembly, all collection chamber components are cleaned with isopropyl alcohol in a horizontal laminar flow bench, except the TEM grids, whose holey carbon film would be destroyed by isopropyl alcohol. Carbon film impurities larger than 0.5 μm are readily characterized in FESEM images.

The method used to produce a complete map of all TEM grids is an automatic scanning procedure, using a FESEM tool that allows choosing an area to scan, at a selected magnification, and a present step of the scan movement. All grids were characterized before the flight. The same areas were scanned with the same resolution when the instrument was returned to the laboratory, in order to locate the collected dust. To minimize contamination, DUSTER was assembled in a clan room. After DUSTER's recovery the Collector and Blank were directly relocated into the FESEM vacuum chamber where it remained until completion of the FESEM scanning procedure.

Collector surface contamination can be indirectly controlled by checking the flight Blank, as this is not exposed to the air flux of stratospheric collection: when a particle that is found on the Collector surface has similar morphology and composition to one on the flight Blank, we can assume that it was not collected in stratosphere. It is from a contamination source.

Ambiguity in stratospheric particles identification can be from: 1) scanning gaps due to a mechanical limit of the FESEM (gaps differ from pre- to post-flight scans, so that post-flight detected in an area corresponding to a pre-flight gap cannot be firmly identified as stratospheric); 2) resolution selection for FESEM scans (0.5 μm). Due to a lack of time to be ready for launch, we had to decide selecting a lower resolution with respect to the DUSTER performance (cut off 0.1 μm).

3. Results and discussion

After recovery of the DUSTER instrument, the post-flight FESEM scan images were compared to pre-flight scan images. In this manner 26 particles were found on the Collector and 3 particles were found on flight Blank. The size distribution of the collected grains is plotted in Fig.1.

DUSTER2008 successfully collected dust with dimensions covering almost three orders of magnitude. More than half of the grains are found in the size range from 0.5 μm to 2 μm; another group lies in the $2 - 10\ \mu m$ range. A large gap is observed from 10 to 100 μm, where only one particle of $\approx 20\ \mu m$ is present; finally, above 100 μm, two very large spheres were collected.

The morphological classification was done using pictures taken by FESEM at low voltage (2 *kV*). The 26 particles can be divided in two

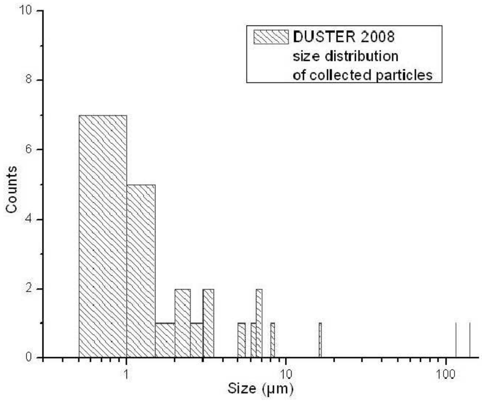

Fig. 1. Size distribution of collected grains.

major morphological classes, spheres (isolated or aggregates) and irregular particles. The two isolated spheres are more than 100 μm in diameter, two aggregates of spherical particles ($\sim 0.5\ \mu m$) (Fig.2). The irregular particles can be divided into three classes using their grain morphology but there is no apparent correlation with particle size:

1. aggregates of irregular grains (Fig.3);
2. fluffy or porous-like aggregates (Fig.4);
3. massive (Fig.5).

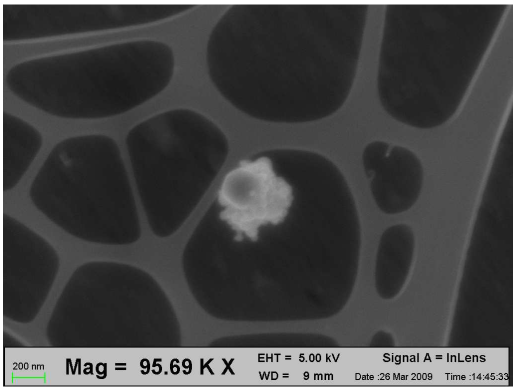

Fig. 2. Particle D08C_031.

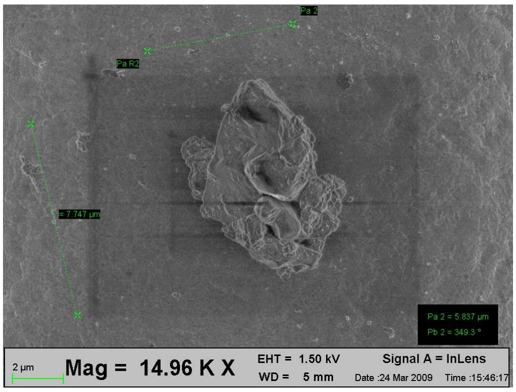

Fig. 3. Particle D08C_006.

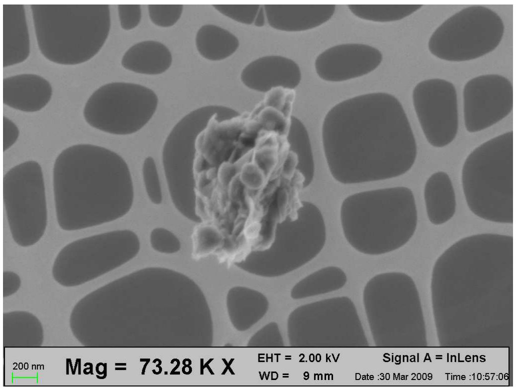

Fig. 4. Particle D08C_017.

Preliminary EDX analyses of the particles was done with a spatial resolution of $0.5 - 1\ \mu m$. For particles larger than 5 μm we were able to perform quantitative analyses. For those smaller than 5 μm we have qualitative analysis. Au and C are the main elements of the TEM grids and the holey carbon thin film. Additional contributions may come from elements present in the stainless steel substrates pins (Palumbo et al. 2008) underneath the TEM grids, (Fe with traces of C, Mn, Si, P, S, Ni, Cr, Mo).

After correcting the quantitative data from contributions by the stainless still substrate (Palumbo et al. 2008), we can divide the particles in two morphological categories: (1) spheres and (2) non-spherical particles that are between 5 and 7 μm (Table 1). The two big spheres contain O, Si, Na, C, Ca, Mg. The particles in the size range $5 - 7\ \mu m$ are rich in Ca, O; two are also rich in F. The presence of fluo-

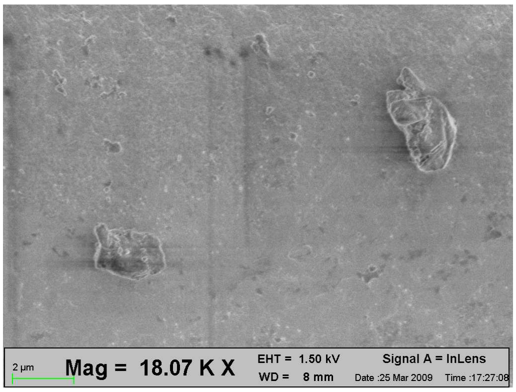

Fig. 5. Particles D08C_015 (left) and D08C_012 (right) are found close to each other and belong to the same morphological class.

Table 1. Quantitative elemental analysis using EDX

Particle	Elements (*weight%*)
D08C_001	O(60.68) Si(26.40) Au(10.04) Na(6.91) Ca(3.56) C(3.37) Mg(1.75)
D08C_002	O(53.50) Si(19.15) Na(7.16) C(6.49) Au(5.23) Ca(2.18) Mg(1.59) Al(0.14)
D08C_006	O(38.65) Ca(28.42) C(21.02) Au(7.28) Fe(3.47)
D08C_007	Ca(31.20) O(29.66) C(19.44) F(11.93) Fe(2.93) Au(2.72) Si(0.47)
D08C_008	Au(48.52) O(25.75) Ca(22.62) C(1.30) Fe(0.71) Na(0.24)
D08C_009	O(47.12) Ca(26.95) C(15.35) Au(9.20) F(2.33)

Table 2. Qualitative elemental analysis using EDX for particles $(1-5)\mu m$ large

Particle	Elements
D08C_003	O, C, Ca, F, Au
D08C_005	O, C, Ca, Fe, Au
D08C_011	Fe, Mn, Ni, Cr, Au Al, K, P, Si, C, Na
D08C_012	O, Ca, Au, C, Mg, Fe
D08C_014	O, Cr, C, O, Fe, Ni
D08C_015	Au, Ca, C, O
D08C_017	C, O, Mn, Cr, Fe, Ni, Si, Au
D08C_021	Au, O, Ca, C, F
D08C_022	Fe, Ni, Mn, Cr, O, C, Si, Au
D08C_023	Au, Fe, Cr, C, O
D08C_024	Fe, Ni, Cr, C, O
D08C_025	Fe, Ni, Au, Cr, O, C

rine is also detected by a qualitative analysis of another larger 20 μm particle; two smaller particles may come from this large particle as they are all in close proximity on the substrate. The qualitative EDX analyses of particles smaller than 4 μm show only elements present in the substrate (Table 2). Relocation of these particles, and further analysis with other instruments will be necessary to identify their origins.

For particles smaller than 3 μm we have insufficient data to constrain their compositions and possible origins. The two large spheres ($>$ 100 μm) have similar O-Si-Na-Mg-Ca (ranked in decreasing abundances) compositions and both show a very smooth surface. These properties suggest they are condensed-liquid spheres such as coal fly ash (McKerall et al. 1982), volcanic ejecta, or perhaps extremely rare micrometeorite spheres due to the ablation of Si-rich interplanetary dust grains (Rietmeijer 2002). These spheres highlight the challenges that are inherent to our type of research. That is, the presence of the Longyearbyen coal power plant near to the balloon launch site suggests that it is a possible, or even likely, source of these spheres. For DUSTER to collect them at its cruising altitude they would have to be hollow coal fly ash spheres, which is not unlikely. Should they be massive spheres the likelihood of an extraterrestrial origin becomes an option. It is an example of how the physical properties will have to be considered when assigning dust particle sources. We will have to determine the hollow or massive nature of these spheres.

The particle in Fig.6 is an almost equant mineral grain that on one side is partially corroded into the highly irregular structure of the left-side. Elemental composition suggests it could be a F-bearing carbonate mineral grain.

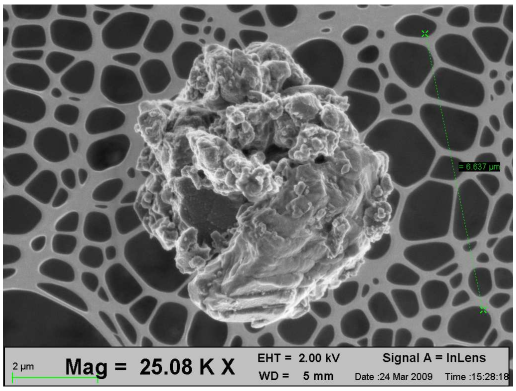

Fig. 6. Particle D08C_007.

4. Conclusions

From the initial analysis of the first DUSTER scientific flight we can conclude that the instrument is capable of collecting solid stratospheric particles in the size range 0.5 – 200 μm at high altitudes. It was also demonstrated that contamination is not a problem during collection and post-flight handling of the samples. The preliminary results of our laboratory analyses show that on DUSTER's first flight is successfully sampled natural and anthropogenic terrestrial dust, and perhaps rare extraterrestrial dust in a size range that was poorly studied so far.

Acknowledgements. The DUSTER project has been funded by the Italian Space Agency, Regione Campania and MIUR (Ministero dell'Istruzione dell'Universit e della Ricerca). We thanks ASI and ISTAR (International Science Technology And Research) for the support given during the operational flight performed within launching site verification activities.

References

Della Corte, V. et al. 2011, Memorie della Società Astronomica Italiana Supplementi, 16, 14

Deshler T. et al. 2003. Journal of Geophysical Research, Vol.108, No.D5, 4167.

Divine, N. (1993. J. Geophys. Res., 98(E9), 17029.

Flynn, G.J. 1997. Lunar and Planetary Institute Science Conference Abstracts, 28, 361.

Grun E., B.A.S. Gustafson, S.F. Dermott & H. Fechtig 2001, (Eds.), Springer.

Junge, C. E. et al. 1961. Journal of Meteorology, Vol.18, 81.

Kasai T. et al. 2003. Review of Scientific Instrument, Vol.74, No.2, 1082.

McKerall, W.C., et al. 1982. Analysis of Fly Ashes Produced in Texas. Texas Transportation Institute, Research Report No. 240-1, Texas A&M University, College Station, Texas.

Palumbo, P. 2008. Memorie della Società Astronomica Italiana, Vol.79, 853.

Rietmeijer, F.J.M. 1993. J. Volc. Geothermal Res. Vol.55, 69-83.

Rietmeijer, F.J.M. 1998. Interplanetary dust particles. In: Papike, J.J. (Ed.), Planetary Materials, Reviews in Mineralogy, vol. 36. Mineralogical Society of America, Chantilly, pp. 2-1 - 2-95.

Rietmeijer, F.J.M. 2002. Chem. Erde 62, 1-45.

Testa J.P. et al., 1990, Earth and Planet. Sci. Letters, Vol. 98, 287-302.

Warren, J.L., Zolensky, M.E., 1994. Collection and curation of interplanetary dust particles recovered from the stratosphere. In: Zolensky, M.E., Wilson, T.L., Rietmeijer, F.J.M., Flynn, G.J. (Eds.), Analysis of Interplanetary Dust, AIP Conf. Proc., vol. 310. American Institute of Physics, New York, pp.105-114.