SATELLITE-BASED CLIMATOLOGY OF AEROSOL COMPONENTS

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ABSTRACT

A synergetic aerosol retrieval method (SYNAER), which exploits a spectrometer radiometer combination to retrieve aerosol optical thickness and type, has been developed and validated at DLR-DFD with GOME and ATSR-2, both onboard ERS-2 (Holzer-Popp et al., 2002a and 2002b). The methodology selects the most plausible type of aerosols from the remote sensing observations in each pixel without relying on any additional background data set. This method is currently adapted to ENVISAT SCIAMACHY and AATSR and will also be applicable to METOP GOME-2 and AVHRR. Furthermore its possible adaption to the synergetic observation of aerosol diurnal variations with TERRA/AQUA-MODIS and MSG-SEVIRI will be studied.

Based on 14 months of data in 1997/1998 from GOME and ATSR-2 over the MSG observation area (due to GOME operations modes only for 3 days each month) a first climatological 5x5 degree data set has been derived, which describes annual average optical thickness values of major components (sulfate/nitrate, soot, dust, sea salt) of the atmospheric aerosol load. The first validation of the method and recent investigations in the community have led to the definition of promising improvements in the aerosol model: Absorption features of soot and mineral components as well as the vertical profile of dust outbreak events require updating. The impact of these changes on the retrieval results will be analyzed in the near future after reprocessing of the climatological dataset has been completed.

The paper summarizes the first preliminary results of this climatology effort (using the old aerosol model without above improvements) and analysis its potential and limitations (in particular due to the restricted data amount) as well as the possibilities to extract seasonal datasets. A reprocessing of the entire dataset with the improved aerosol model is currently under way. Application of the methodology used in this paper with pairs of different satellite instruments on polar orbiting and geostationary platforms holds large potential for several application areas: long-term <u>climatology</u> of aerosol components, daily observation of different particles as important feature of <u>chemical weather</u>, and the <u>assimilation</u> of aerosol components into a hemispheric air quality forecast model.

1. INTRODUCTION

Air pollution by solid and liquid particles suspended in the air, so-called aerosols is one of the major concerns in many countries on the globe. One focus of concern is related to aerosols from anthropogenic origin mainly by combustion processes (industry, vehicle transport, heating, biomass burning). In developed countries improved combustion and filtering methods have led to a general decrease of particle concentrations in total suspended matter but new concern arises from potential health impact of increasing numbers of smaller aerosols, so-called nano-particles in particular from diesel engines (Pope, III et al 2002). Stedman (2004) reports that 21 – 38 % of total excess deaths in the UK during the summer heatwave of 2003 are attributable to elevated ozone and particle concentrations. On the other hand developing countries suffer still from high total particle loads in the air. Furthermore, natural aerosols (mainly dust and sea salt) also contribute significantly to background and episodically severly increased particle concentrations. Dust can also act as carrier for long-range transport of deseases, e.g. from the Sahara to the Caribic or Western Europe (Pohl 2003), or even once around the globe (Prospero et al. 2002), as recently Chinese yellow sand was detected as far as the Swiss Alps. Also well known in principle are direct (by reflecting light back to space) and indirect (by acting as cloud condensation nuclei) climate effect of aerosols, although large uncertainties exist in the exact values of the forcing. Recent publications (e.g. Petzold et al., 2004) point out, that the absorption behaviour of particles (mainly soot and minerals) needs to be known to assess their total climate effect (strongly absorbing particles can regionally reverse the sign of the aerosol forcing from cooling to heating).

In the light of this overall picture, climate monitoring, long-term air pollution monitoring as well as short term forecast of pollution levels need to take into account intercontinental transport processes and the composition of the atmospheric particle load. Satellite observations of the total aerosol mass have experienced significant improvements in the last few years thanks to improved instrumentation and enhanced retrieval algorithms (Kaufman, et al. 2002). Thus they offer the potential to regularly monitor the global aerosol distribution and by assimilating these measurements into chemistry-transport-models to enhance particle forecasts especially for episodic severe pollution events. Furthermore, satellite data can contribute to deduce background and long-range transport patterns of aerosols. And finally, with the emerging capacity to separate the total aerosol mass observed with optical observations into major components, a better understanding of the behaviour of different chemical and size classes is supported. As a first step towards this goal a satellite-based climatological data set of the optical thickness of major aerosol components over Europe / Africa is presented here which can be used as background aerosol distribution maps.

After a brief introduction in section 1 the retrieval method as well as the status of validation efforts against AERONET spectral sun photometer measurements and other satellite observations are described in section 2. Section 3 deals with the improvements in the aerosol model and section 4 describes the first preliminary results of the application of the synergetic method to the 14 month dataset over Europe and Africa. Sections 5 to 7 contain the summary, acknowledgements and references.

2. RETRIEVAL METHOD: OVERVIEW AND ITS VALIDATION STATUS

At the German Remote Sensing Data Center (DFD) the new aerosol retrieval method SYNAER (SYNergetic AErosol Retrieval) was developed (Holzer-Popp et al. 2002a) which delivers boundary layer aerosol optical thickness (BLAOT) and type over both land and ocean, the latter as percentage contribution of 6 representative components from the OPAC (Optical Parameters of Aerosols and Clouds, Hess et al., 1998) dataset to BLAOT. The high spatial resolution of the ATSR-2 instrument (Along Track Scanning Radiometer) permits accurate cloud detection, BLAOT calculation over automatically selected and characterized dark pixels and surface albedo correction for a set of 40 different pre-defined boundary layer aerosol mixtures. After spatial integration to the larger pixels of the spectrometer GOME (Global Ozone Monitoring Experiment) these parameters are used to simulate GOME spectra for the same set of different aerosol mixtures. A least square fit of these spectra to the measured spectrum delivers the correct BLAOT value and - if a uniqueness test is passed - the aerosol mixture. For humidity dependent components two models with 50% and 80% relative humidity have been included. SYNAER is currently under implementation for operational processing at the German Remote Sensing Data Center, which also serves as processing and archiving center (PAC) for all atmospheric ENVISAT data on behalf of ESA.

Accurate cloud detection is an important prerequisite for each aerosol retrieval. The well established APOLLO (AVHRR Processing Scheme Over CLoud Land and Ocean; Kriebel et al. 2003) software was adapted to ATSR-2 data. The capability of retrieving cloud cover in boxes of 1 km^2 means a significant strength of SYNAER because it reduces the erroneous aerosol detection due to the presence of sub pixel clouds significantly. It even allows the correction of partly cloudy GOME pixels. First case studies distributed over the globe using ground based sun-photometer measurements of the spectral aerosol optical thickness from NASA's Aerosol Robotic Network (AERONET) at 14 locations and one airborne lidar measurement show a good agreement with errors less than 0.1 in six wavelengths (340 – 870 nm) which indicates correct assessment of the amount and type (namely the spectral dependence of extinction) of aerosol (Holzer-Popp et al. 2002b). This first groundbased validation comprised data from 4 continents in several climate zones (latitudes 17 South to 56 North) distributed over 2 years except the winter season with solar elevations ranging from 25 to 60 degrees. The cloud fraction inside the GOME pixels ranged up to 35%. Furthermore, a comparison of monthly mean results from SYNAER and other satellite aerosol retrievals as well as AERONET stations over ocean (Myhre et al., 2004) showed a satisfactory agreement with the other datasets.

3. IMPROVEMENTS IN THE AEROSOL MODEL

Basically the components (which summarize the optical features of all particles with similar optical behaviour) including their log-normal size distribution are taken from the OPAC database (Hess et al., 1998). Table 1 summarizes their relevant microphysical properties and derived (Mie-calculated) optical characteristics. However, more recent campaigns and AERONET data exploitations have been used to improve some specific items:

- the original soot component was split in 2 components for weakly absorbing biomass burning (Dubovik, 2002) and very strongly absorbing diesel (industrial) carbon (Schnaiter, et al., 2003). Furthermore, the real part of the refractive index was adjusted to Dubovik, 2002 representing a mixture of Amazonian, South American cerrado, African savannah and boreal forest fires.
- in the absorption of mineral dust (transported and insoluble) 2 components were introduced to take the dust origin with different hematite content and consecutively absorption into account (Dubovik, 2002 representing an average of Cape Verde, Saudi Arabia and Bahrain observations and Sinyuk et al., 2003 below 400 nm). In the case of desert outbreak (transported minerals) the lowest aerosol layer of 4-6 km was modeled as two distinct sub-layers (dust above background), as they occur in nature.

Component	Species	Complex refract. Index at 550 nm	Mode radius [µm]	Stand. Dev. of size distribution	Particle density [g/cm^3]	Extinction coefficient for 1 particle per cm^3 at 550 nm [km^{-1}]	Single scattering albedo At 550 nm	Literature source
WASO, rH = 70%	Sulfate/nitrate	1.53 – 0.0055 i	0.028	2.24	1.33	7.9 e-6	0.981	Hess, et al. 1998
INSO	Mineral dust, high hematite content	1.53 – 0.008 i	0.471	2.51	2.0	8.5 e-3	0.73	Hess, et al. 1998
INSL	Mineral dust , low hematite content	1.53 – 0.0019 i	0.471	2.51	2.0	8.5 e-3	0.891	Dubovik, et al. 2002
SSAM, rH = 70%	Sea salt, accumulation mode	1.49 – 0 i	0.378	2.03	1.2	3.14 e-3	1.0	Hess, et al. 1998
SSCM, rH = 70%	Sea salt, coarse mode	1.49 – 0 i	3.17	2.03	1.2	1.8 e-1	1.0	Hess, et al. 1998
BISO	Biomass burning soot	1.63 - 0.036 i	0.0118	2.0	1.0	1.5 e-7	0.698	Dubovik, et al. 2002
DISO	Diesel soot	1.49 – 0.67 i	0.0118	2.0	1.0	7.8 e-7	0.125	Schnaiter, et al. 2003
MITR	Transported minerals, high hematite content	1.53 – 0.0055 i	0.5	2.2	2.6	5.86 e-3	0.837	Hess, et al. 1998
MILO	Transported minerals, low hematite	1.53 – 0.0019 i	0.5	2.2	2.6	5.86 e-3	0.93	Dubovik, et al. 2002

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1. Table: updated aerosol components (new components are highlighted)

Table 2 shows the (updated) definition of the 40 mixtures used in the SYNAER retrieval method. Values in the table show the vertical profile, relative humidity in the boundary layer and the percentage contribution to the optical thickness at 550 nm of the respective components. Two groups of 20 mixtures, each are applied where either relative humidity or the hematite content of the mineral component (and consecutively the absorption) are altered. Alternative values are marked with grey boxes in table 2: For example, mixture number 1 has 50% relative humidity and mixture number 21 has 80% relative humidity; mixture number 2 has 5% insoluble (high hematite content) component, whereas mixture number 22 has 5% insoluble (low hematite content) contribution to the optical thickness at 550 nm. The set of 40 mixtures is meant to model all principally existing aerosol types and allow for some variability of the composition of each type. These set of mixtures has proven to provide a fit in the GOME spectra retrieval which is in many cases better than a 1% noise level. Work to determine the success rate of the retrieval approach using this set of mixtures with a 14 month dataset is currently going on. In the old SYNAER aerosol model the (one) OPAC soot and mineral component was used.

					Component contributions to AOT550 [%]								
No.		**Name**	**Rel. hum. [%]**	**Vert. Prof. [km]**	**WASO**	**INSO**	**INSL**	**SSAM**	**SSCM**	**BISO**	**DISO**	**MITR**	**MILO**
1	21	Pure watersoluble	50/80	2	100								
2	22	Continental	50	2	95	5	5						
3	23				90	10	10						
4	24				85	15	15						
5	25	Maritime	50/80	2	30			70					
6	26				30			65	5				
7	27				15			85					
8	28				15			75	10				
9	29	Polluted watersoluble	50/80	2	90						10		
10	30				80						20		
11	31	Polluted Continental	50	2	80	10	10				10		
12	32				70	10	10				20		
13	33	Polluted Maritime	50/80	2	40			45	5		10		
14	34				30			40	10		20		
15	35	Desert Outbreak	50	2 - 4	25							75	75
16	36			3 - 5	25							75	75
17	37			4 - 6	25							75	75
18	38	Biomass Burning	50/80	3	85					15			
19	39				70					30			
20	40				55					45			

WASO = watersoluble, INSO = insoluble, INSL = insoluble / low hematite, SSAM = sea salt accummul. mode, SSCM = sea salt coarse mode, BISO = biomass burning soot, DISO = diesel soot, MITR = mineral transported, MILO = mineral transported / low hematite; Mixture number N and mixture number N + 20: alternative humidity or mineral composition, respectively

2. Table: updated aerosol mixtures

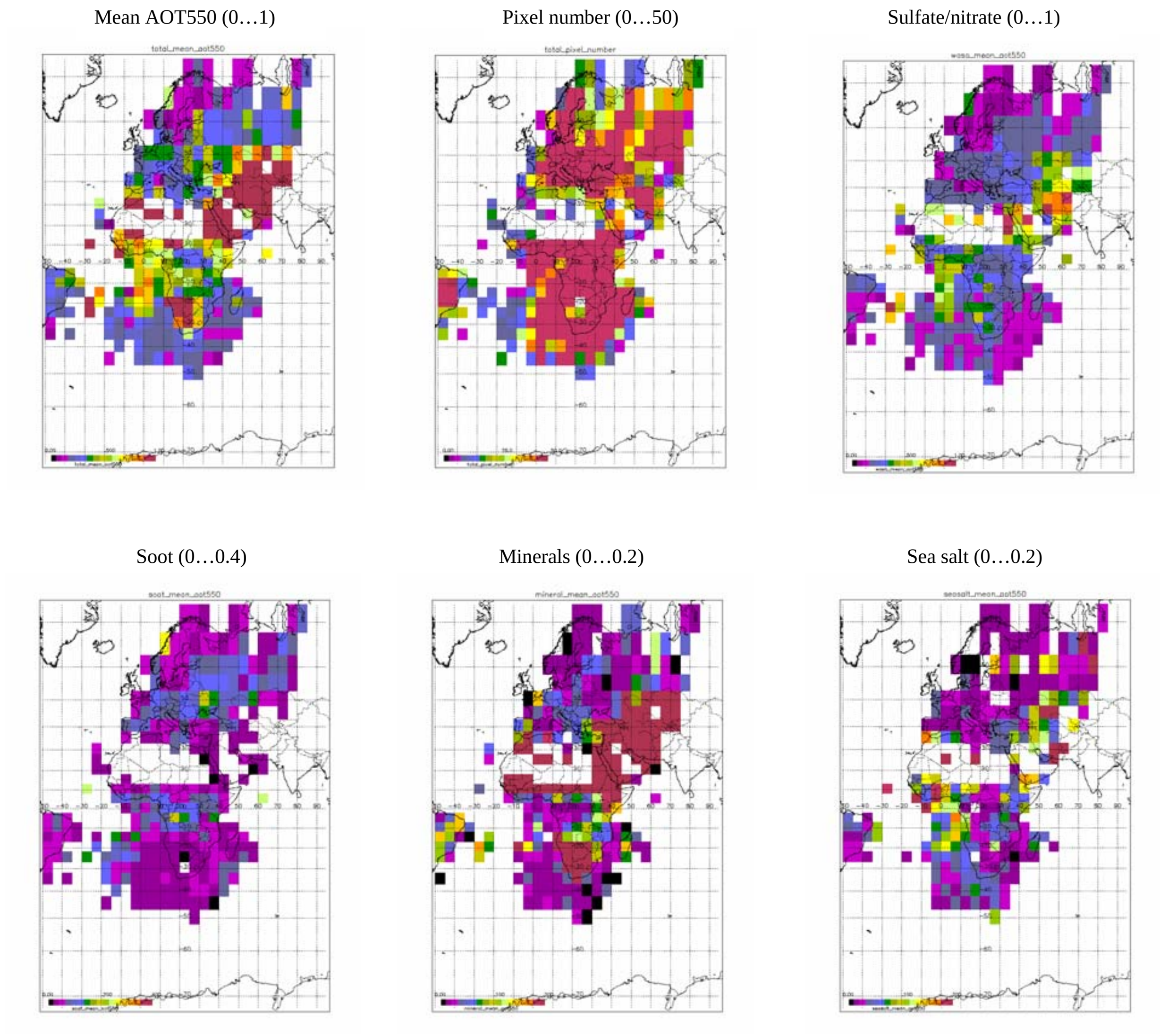
4. FIRST PRELIMINARY 14-MONTH CLIMATOLOGICAL DATASET

It is the ultimate goal of this work to produce (and in future update) a satellite based aerosol climatology for the observation area of the European geostationary Meteosat Second Generation satellite. For this purpose the SYNAER method will be implemented and work operationally with the sensors SCIAMACHY and AATSR onboard ENVISAT. A backup climatology production based on GOME and ATSR-2 products (both onboard ERS-2) of the year 1997/98 is shown here. The final aim is to deliver 4 seasonal climatology datasets with a 1 degree horizontal grid. Due to cloud coverage and method inherent limitations a one year dataset was obtained as first product for a 5 degree grid.

For this first application of the SYNAER method data products of the period July 1997 through August 98 covering Europe/Africa were received through the ESA AO project SENECA (AO ID-106). Unfortunately, GOME measures “small” pixels of 80x40 km^2 only for 3 days every month, and 320x40 km^2 pixels throughout the rest of the month. For producing the aerosol climatology only “small” pixels are meaningful in corresponcance with horizontal variability scales of the aerosol loading and are thus exploited. With this small pixel mode GOME covers a swath width of 240 km with only 3 pixels in one scan line (SCIAMACHY will deliver pixels of 60x30 km^2 for a swath of 960 km, i. e. 16 pixels in one line). Although this means a severe limitation to the available data base, it opens the opportunity to test the methodology with a one year dataset. Detailed handling of quality information from the retrieval process such as fit error, GOME-ATSR-2 cross-calibration deviation, spectral noise, surface elevation, solar elevation angle, etc. was optimized. Depending on the cloud coverage and method inherent limitations (surface brightness must not exceed 8% over land and 1.5 % over ocean, GOME pixel cloud fraction must not exceed 35%, differentiation of aerosol types is only reliable for optical thickness at 550 nm larger than 0.1, the ambiguity test rejects pixels with a fit error less than 0.005) extracting the seasonality was hardly possible with this first dataset. Given the limited temporal coverage of 3 observations per month (which is further reduced due to cloudiness and technical errors) it was assured during averaging of the climatology values that a minimum of 2 orbits (i.e. 2 different times of observations) and of 5 pixels contributed to the box average (mean values for grid boxes with result values are 6 orbits and 41 pixels). To obtain a significant result a 14 month dataset (average and standard deviation optical thickness, number of pixels contributing to one grid box, differentiation of optical thickness into basic components) using a total number of 18914 GOME pixels was produced as first product for a 5 degree grid.

Figure 1 shows the climatology dataset based on the 14 months of GOME/ATSR-2 data on a 5 degree grid. The low number of observations in some boxes (particularily in the South-West Atlantic and at the edges of the dataset) leads to some unrealistic mean concentrations values which are determined by single episodes. This will be enhanced by using the daily ENVISAT observations. However, despite of the small data base the aerosol distribution is rather smooth with a mean aerosol optical thickness of 0.26 and some interesting features can already be observed: Largest optical thickness values above 1 occur over / near the desert areas, whereas the Scandinavian area or oceanic zones far off from the continents show lowest values. Also biomass burning plumes from South America and Central Southern Africa are indicated over the Atlantic.

Figure 1 shows also the component-wise mean aerosol optical thickness maps: Sulfate/nitrate aerosols which are included in all modelled aerosol types as background contribution show even clearer the unpolluted oceanic and Northern areas. Soot occurs most prominently over industrialized / densely populated areas in Central/Eastern Europe as well as over biomass burning source areas in Central/Southern Africa and in their plumes over the ocean. Maximum 14 month 5 degree box average values are near 0.4. Few grid boxes show exceptionally high values which are due to a low number of contributing pixels. Dust is dominant in and around desertic areas (Sahara, Namib, Near East) and occurs with smaller amount in continental and oceanic aerosol. Sea salt occurs with low values also at inland locations which indicates to the limits of separating this component with low impact on the total optical thickness, but it should be noted that its maximum occurs over the Southern Atlantic. Some areas are not covered by the basic dataset because of either too high cloudiness or too bright surface. Due to miss-interpretation because of the simplified dust vertical structure high soot and sea salt values occur over deserts to some extent. Furthermore, an error in the land sea mask was found in the South-East-Atlantic which leads to some increased concentration levels over the ocean. Finally, new ground-based measurements revealed limitations in the soot and dust representation in the SYNAER aerosol model (see section 3), which are also a potential reason of wrong aerosol type detection.



1. Figure: climatology of aerosol components

5. SUMMARY AND CONCLUSION

The potential of the SYNAER method for separating basic aerosol components has been demonstrated with a limited 14 month database. The method will be optimized (especially with regard to absorbing aerosl components) and used for the production of an aerosol component climatology from space. For this purpose SYNAER will be adapted to ENVISAT SCIAMACHY and AATSR by the end of 2004 to produce daily global aerosol maps with 480 km swath and 60x30 km^2 pixel size. Furthermore, simplifications in the current method setup, which are responsible for miss-selection of the aerosol type in certain conditions are currently revised to lead to a more accurate optical thickness retrieval and differentiation of the aerosol type. Then, a reprocessing of the entire GOME/ATSR-2 background 14 month climatology dataset will be conducted, which takes these improvements into account. The land sea mask error will be corrected.

6. ACKNOWLEDGEMENT

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