Aerosol modeling

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1. Introduction

The presentation intends to present recent advances and examples on three questions: a) Why should we care about the aerosol? b) How should we represent the aerosol in a global model? And c) What is the current model quality with respect to observations? The work presented is not intended to provide a review on these topics but assembles hopefully illustrative examples. It is based on recent work in the framework of the GEMS-AER project, where an aerosol module is introduced into the IFS model system at ECMWF with the project partners; it also reflects the parameterisation work being undertaken at the LSCE/IPSL to introduce a cpu-efficient aerosol module in the IPSL coupled climate model and finally builds largely on the experience in the international AeroCom aerosol model intercomparison initiative.

2. Why should we care about the aerosol?

Aerosols modify the Earth radiation budget, but the direct radiative forcing estimate is still associated with a large error bar [*IPCC*, 2007], which calls for a better quantification. The direct aerosol effect is estimated at -0,5W.m^{-2} (-0,9 to -0,1) and satellites can provide an improved understanding of the radiative fluxes [*Bellouin et al.*, 2005; *Kaufman et al.*, 2005]. As an example of significant source of uncertainty one may recall that above clouds the direct forcing by anthropogenic aerosols can be positive in case of the presence of absorbing aerosols such as biomass burning smoke [*Schulz et al.*, 2006]. The interaction of aerosols with clouds is generally suggested to be even more uncertain than the direct radiative forcing.

The radiative forcing by anthropogenic aerosols can be suspected to have masked and obscured the climate effect of the continuous rise of long lived greenhouse gas concentrations (LLGHG) in the last century [*Crutzen et al.*, 2003]. This obscurity raises scientific questions about the quantification of the aerosol effect [*Anderson et al.*, 2003]. Even in times where the anthropogenic cause for climate change is considered virtually certain [*IPCC*, 2007] answering these questions has considerable impact on climate mitigation policy choices. Due to the short life time of suspended particles and eventually more efficient air pollution control the aerosol effect might become relatively small in the future as compared to that of the long lived greenhouse gases (LLGHG), where the effect is proportional to the accumulated emissions [*Dufresne et al.*, 2005]. However, understanding the global aerosol effect is crucial to predict the future evolution of climate, since prediction of future change builds on understanding the past for which observations exist and it will become crucial if greenhouse gas emissions become effectively controlled. While it can be stated that the positive radiative forcing of greenhouse gases is well quantified, the major negative radiative forcing by aerosols is much more uncertain [*Forster et al.*, 2007]. The climate sensitivity of the Earth system, which is defined as the relation of total forcing and global surface temperature increase, owes considerable

uncertainty to the less well defined aerosol forcing. High climate sensitivity would result in a proportionally warmer climate system for a given amount of LLGHG emissions. With respect to future climate pathways two possibilities appear: First, if we have overestimated the recent aerosol effect, then the observed changes in the climate system are already now almost solely due to the increases in LLGHG concentrations. Further increases in LLGHG emissions would provoke a direct and as intense reaction of the climate system probably well described as the lower bound of the IPCC climate model predictions. Alternatively, if we have underestimated the recent aerosol effect, then any reduction in aerosol loads such as air pollution abatement measures or simply reduced emissions would have a large impact on the climate system evolution. The climate sensitivity would be in the upper end of the estimated range of 2°-4.5° per doubling of CO_2 concentrations. Since the aerosol is short-lived as compared to the LLGHG, any future reduction in anthropogenic aerosol loads would reduce the negative radiative forcing components, with the notable exception of the uncertainty in the climate warming effect of black carbon. The LLGHG forcing would be less masked and would be very effective to rapidly change a sensitive Earth climate. Air pollution control of particulate matter would become a source of climate warming [*Andreae et al.*, 2005].

The link between air quality and climate change related policy questions makes research on aerosols an exciting applied science, while the difficult underlying scientific questions make it a field of fundamental research. Investigating the radiative forcing by aerosols requires studies which explore global properties of the aerosol based on highly variable concentrations and distributions and studies which integrate the nature and structure of omnipresent super- and sub-micron particles. Compared to globally 342 Wm^{-2} incoming solar radiation only 0.5 Wm^{-2} is reflected back to space directly by anthropogenic aerosols. Do we understand the global aerosol well enough to predict such a relatively tiny aerosol radiative effect?

3. How to represent the aerosol in a global model?

Aerosol particles may travel varying distances from centimetres to around-the-world [*Damoah et al.*, 2004]. The average life time of an aerosol particle is of the order of days in the troposphere. Aerosol belongs thus to the short-lived atmospheric constituents. The properties of the particles are a function of the physical and chemical environment surrounding them, but particles also have a significant influence on the atmospheric environment by modifying radiation fluxes and taking part in chemical reactions. Several different inorganic and organic chemical species are recognised as the major constituents of the aerosol and are distributed on particles of size ranging from nanometers to several micrometers. Finally, the spatial heterogeneity of the emission sources for primary and secondary aerosol particles is at the origin of the global aerosol field. Mass and number fields of the different species in consequence appear as a fast moving and changing global patchwork. These properties of the aerosol are responsible for significant effects on human health, transport of nutrients and toxic substances and regional to global effects for the radiative balance.

A global aerosol model has to balance the completeness of the aerosol description and the available computing resources. Aerosol modelling is a typical example, where highly parameterised functions are used to describe the way several complex processes are coupled in nature. A cpu-efficient model allows to reject or to adopt a hypothesis much faster, e.g. using repeated numerical experiments with slightly modified input parameters. This also facilitates evaluation of the model with observations through simulations spanning larger periods. Aerosol measurements are widely dispersed in time and space, mainly because they have been obtained mostly during intensive, but scattered campaigns.

Even though any effect of the aerosol, on e.g. the radiation balance, is linked to first order to the particulate matter concentration itself, other aerosol properties need to be described to link cause and effect. These aerosol properties are highly variable in time and space and can be explained by various states of aerosol

mixing [*Raes*, 2000]. This is a major difference from reactive gases where e.g. the radiative properties are almost constant in the atmosphere. Aerosol particle populations are transformed in multiple ways during transport. We can identify the following major characteristics of the ambient aerosol:

- A co-existence of several particle populations from different sources with different size parameters, spanning several orders of magnitude in size, is widely observed. The size distributions of these populations are to first order lognormal.
- Two types of submicron particle populations can be differentiated, of which one population is mostly responsible for the hygroscopicity of the aerosol.
- The fine particles which are found to be less susceptible to water uptake are thought to be composed of organic matter and soot.
- The hygroscopic submicron population is generally considered to consist of internally mixed particles, which may contain varying fractions of sulphate, nitrate, ammonium, sea salt, organic matter and even black carbon.
- Organic matter and black carbon is believed to undergo rather rapid ageing processes, which transform insoluble primary organic and soot particles into soluble ones.
- Dust particles are to be found at distinctly larger sizes than submicron particles, which supports the predominantly external mixing of these particles with the submicron particle population. However, heterogeneous reaction on dust surfaces may add nitrate and sulphate to dust particles.
- Similarly, sea salt is found at larger diameters, but may contain nitrate and sulphate from condensation of reactive gases on its surface. The variety of sea salt formation processes is probably the reason for a broader sea salt than dust size distribution.
- Temporal evolution of the aerosol size distribution is most important in the small sized Aitken and Nucleation mode, but then happens relatively quickly. These two modes contribute little to aerosol surface and thus optical properties.

Altogether we can assume that four basic properties of the ambient aerosol matter most: *size, chemical composition, hygroscopicity and mixing state of the particles*. There should be enough freedom in the model structure to represent and express the ambient variability in these basic properties.

Size: One possibility to resolve the parameterization problem is the superposition of several lognormal distributions (modes) as an appropriate model structure. Each individual mode may carry slightly different characteristics with respect to size, composition, hygroscopicity and mixing. Each mode is described by one number concentration tracer and several mass concentration tracers. Each aerosol mode belongs to either a soluble (rather hygroscopic) or insoluble category. Such as concept is used for instance in our LMDzT INCA model and also in the ECHAM5 aerosol module [*Stier et al.*, 2005]. As shown by [*Schulz et al.*, 1998], through comparison with an explicit multiple bins scheme for eg the sedimentation of dust, it is possible to follow the size evolution in a computational efficient way, if the width of the distribution is assumed constant and if the width of the distribution is not larger than approximately two in value. The advantage of the modal approach is that only two tracer concentrations are needed to describe the full size spectrum of one mode, e.g. number and total mass. The link of several mass tracers to one number tracer implies that each major aerosol mode is composed of internally mixed aerosol particles, an understanding which is supported by observations. Important parameterisation choices with respect to size are the initial source size

distributions and the way how removal processes effect the size distribution and how different aerosol modes may interact. The size distributions may change by several processes:

- Gas-phase oxidation of SO_2 by OH radicals and any other new particle formation process forms new particles. This process, through aerosol microphysical growth processes, creates varying number and mass in the nucleation, aitken and accumulation mode.
- Aqueous phase production of secondary aerosol mass such as sulphate creates aerosol mass in the accumulation mode. Such production will change the number concentration probably very little even when clouds evaporate again.
- Heterogeneous reactions and condensation of semi volatile substances on the aerosol surface add aerosol tracer mass but no number.
- Size selective wet removal and sedimentation selectively removes the larger particles in each mode, which would shift the mode diameter constantly to smaller diameters.
- Advection and mixing are responsible for the mixing of different aerosol populations. A local size distribution will thus be in constant change.
- Primary, insoluble carbonaceous particles become soluble with time. This ageing process transfers insoluble accumulation mode particle number and mass into the soluble accumulation mode.
- Hygroscopic growth is changing the size of the particles instantaneously as a function of ambient relative humidity. It creates important changes in size and can be e.g. responsible for accelerated sedimentation and optically different diameters.

Chemical composition: A number of chemically distinct components make up the global aerosol. A good assumption is to distinguish at least between dust, sea salt, sulphate and nitrate, black carbon and particulate organic matter. The exact chemical composition of these components might vary because of their internal chemical composition (sulphate e.g. might be present as ammonium sulphate, ammonium bisulphate, gypsum or sodium sulphate). Dust and sea salt are the most important primary, natural aerosols in terms of mass and optical depth. Sulphate is the major part of the anthropogenic aerosol component. Black carbon is strongly light absorbing and dominates the single scattering albedo in polluted and semi-polluted regions. Particulate organic matter is the major product of biogenic and anthropogenic combustion processes. Chemical reactivity of particles may change with time through the interaction with the gas phase and changed surface properties. Particle composition itself may change through interaction with the gas-phase, if for instance sodium chloride in sea salt reacts with nitric acid to form sodium nitrate. Ammonium nitrate and semi-volatile organic compounds may evaporate and condense as a function of temperature and vapour pressure of the volatile substances. All these considerations call for a representation of this chemical composition in an aerosol model through independent tracers. The predominant presence of the species in aerosols of different size can be used to limit the complexity of the tracer representation system.

Hygroscopicity: The uptake and loss of water on aerosol particles is generally fast and depends on the chemical composition, size and surface properties of the aerosol particle. Hygroscopic growth of aerosol particles is reported to be a major factor in explaining the optical parameters of an aerosol population. Several attempts have been made to establish growth factors as a function of composition and size[Swietlicki et al., 2008]. [Swietlicki et al., 1999] have reported that in many instances a co-existence of hydrophobic and hydrophilic particles can be observed. Upon hydration of dry particles of a given diameter two particle

modes with distinct hygroscopic growth factors appear. By differentiating between soluble and insoluble modes in the model structure, such observations can be integrated in the model. The change in overall aerosol hygroscopicity may be followed as a function of contributions from differently hygroscopic aerosol modes. Hygroscopic growth requires to be computed as a function of chemical composition in each mode.

Mixing state of the aerosol: Observations have both shown the existence and even coexistence of externally and internally mixed particles. Primary particle emissions and inefficient particle merging processes are responsible for external particle populations. However, microscopic observations of individual particles and the coherent macroscopic properties of aerosol populations have shown that internally mixed particles are also widespread. Cloud processing, heterogeneous reactions, coagulation, vapour evaporation and vapour condensation are processes that can create internally mixed particles. The mixing state can also change due to ageing of organic particles, due to the formation of coarse nitrate on dust and sea salt and due to the condensation of sulphuric acid on fine particles. The interaction with reactive gases is the principal cause for a change in the chemical composition of the aerosol during transport. Aerosol dynamics, which would transfer aerosol mass from one mode to another one, can be formulated as shown by [Vignati et al., 2004]. In principle it would be possible to formulate within each aerosol mode a specific mixing state if such a description would be desirable. For example: Optical properties of the soluble accumulation mode differ, if one assumes a core of black carbon in all particles or just in 30% of the particles.

Altogether, the interpretation of the aerosol tracer concentrations with respect to the resulting aerosol properties is considerably more flexible, if the tracers are grouped in several lognormal modes. Even if the parameterisations of the aerosol dynamics and the interactions with the gas phase are probably far from well constrained, the modal approach allows to describe the aerosol in a computational efficient way and can be made more complex in the future.

4. Current model quality with respect to observations

Current aerosol model quality has been recently documented e.g. via the AeroCom model intercomparison [*Kinne et al.*, 2006]. New developments are constantly made to complexify the aerosol modules and the description of the aerosol-cloud interactions. Selected additional components have been added such as nitrate and secondary organic particulate matter, as well as ultrafine and mixed particles. At the same time it is still not clear why the models are as diverse as they are with respect to aerosol life time, removal processes, optical properties and forcing estimate (see for instance discussion in [*Textor et al.*, 2007]). A better understanding of this model diversity would allow identifying those crucial processes which need improved parameterization and observational constraints. Figure 1 illustrates as an example the differences in simulated aerosol optical depth at 550 nm (AOD) at four typical sites where one aerosol type is dominating. As a reference the AeroCom Median model is shown, constructed from the individual models, plotted in the background. Simulation results are compared to sun photometer derived aerosol optical depth from Aeronet. Significant systematic biases can be detected which often depend on season.

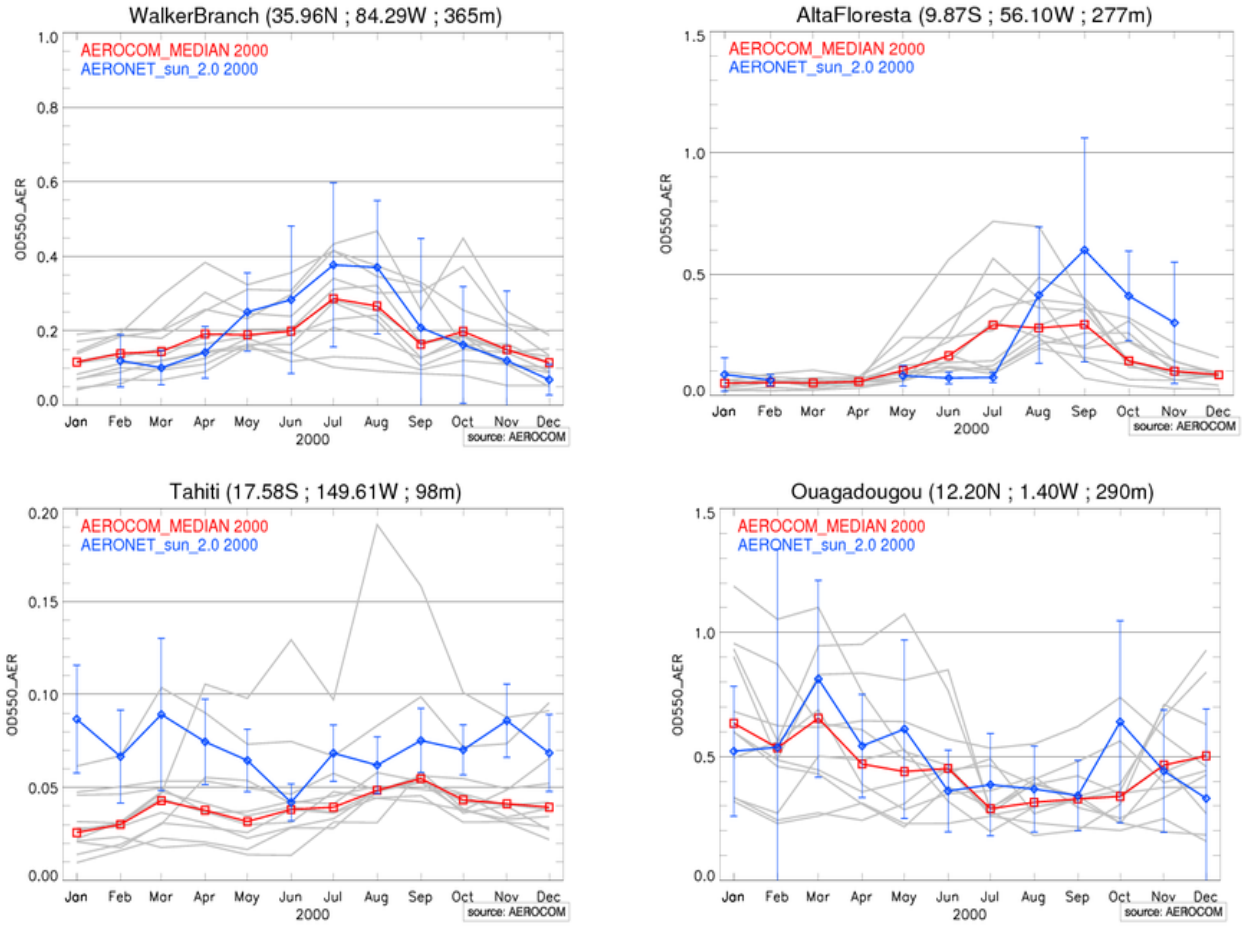

Figure 1 Aerosol optical depth at four sites characteristic for pollution, biomass burning, sea salt and dust. AeroCom A models are shown in grey, the AeroCom Median model in red and the Aeronet sun photometer data in blue. Stations are typical for pollution (Walker Branch), biomass burning (Alta Foresta), marine aerosol (Tahiti) and dust (Ouagadougou)

4.1. Evaluation of experimental LMDzT-INCA dust forecast system

One example for the extent to which we can trust nowadays global aerosol model is our own global aerosol transport model LMDzT-INCA which is operated for an experimental chemical weather forecast system. The model includes a dust aerosol component as part of a multi-component aerosol module [*Balkanski et al.*, 2004]. For the forecast version winds are nudged, which means that three dimensional wind fields for every 6 hour time step for all the forecasted time period are used from NCEP and ECMWF to guide the model wind fields and hence force the atmospheric model LMDzT to follow synoptic weather evolution. For experimental purposes we use three day forecasted winds for NCEP and five days from ECMWF. We use the standard LMDzT-INCA model as described e.g. by *[Hauglustaine et al., 2004]* and its aerosol module as documented within the AeroCom model intercomparison effort *[Textor et al., 2006]*.

Dust erosion by wind is responsible for the largest fraction of the global aerosol mass load. Erosion takes place mainly in semi-arid and arid regions but not everywhere in such regions and not at all times. While the erosion of dust is a strong function of wind speed, approximately to the power of three, it is necessary to account in a dust source model for limitations in erodibility. We have implemented several such limitations: the "*erosion potential*" of a given region provides the maximum amount of dust which is effectively available for atmospheric transport under favourable wind speed conditions; the "*varying state of the soil*" can introduce a variability in the erosion potential through wetting of the soil and freezing conditions; the

"*threshold wind speed*" expresses that limit above which wind stress can effectively start the erosion process; the initial "*dust size distribution*" corresponds to that fraction of the saltated soil particle spectrum which is long range transported. The simplified formula of the INCA dust flux F_{dust} reads as follows:

$$F_{dust} = C * u^2 * (u-u_t) * F_{soil} \quad (\text{if } u > u_t)$$

where C represents the erosion potential, u the surface wind speed, u_t the threshold velocity above which wind erosion can take place, F_{soil} represents the inhibiting factor due to unfavorable soil conditions. The erosion potential C is varied for different dust minerals and for number and mass of uplifted dust to provide an initial size distribution.

The experimental forecast system is running since summer 2005. It has been originally built to support experimental campaigns. Each daily forecast simulation encompasses a period of three days (based on NCEP winds) or five days (based on ECMWF winds). While the nudging is done at the model resolution of 3.75x2.85 degrees, forecasted surface winds are used in addition at a higher resolution of 1x1 degree to compute with higher accuracy the dust fluxes. The results of the forecast model with respect to the evolution of the aerosol and gas species are accessible via a web interface: http://www-lsceinca.cea.fr/cgi-bin/lsce/inca_work_annualrs.pl. A reanalysis run was performed later which made use of the operational ECMWF winds.

The comparison of the daily time variation of AOD is shown exemplary for the Aeronet/Photons site near Dakar in Figure 2. Despite its coarse resolution, the forecast model captures 48% of the day to day dust variability within the year 2006 near Dakar (reanalysis model version 50%). On weekly and monthly scale the model captures respectively 63% and 71% of the variability (reanalysis version: 70% and 74%). Most of the smaller dust events are found both in the Aeronet and modeled time series. For some large dust events the coherence is less well. We explore also the deterioration of dust forecast with time of forecast. Correlation coefficients between daily AOD values observed and modelled at Dakar decrease from 0.69 for the initial forecast day J0 to 0.59 and 0.41 respectively for two days ahead and five days ahead. The correlation found for the reanalysis run is found to attain 0.71.

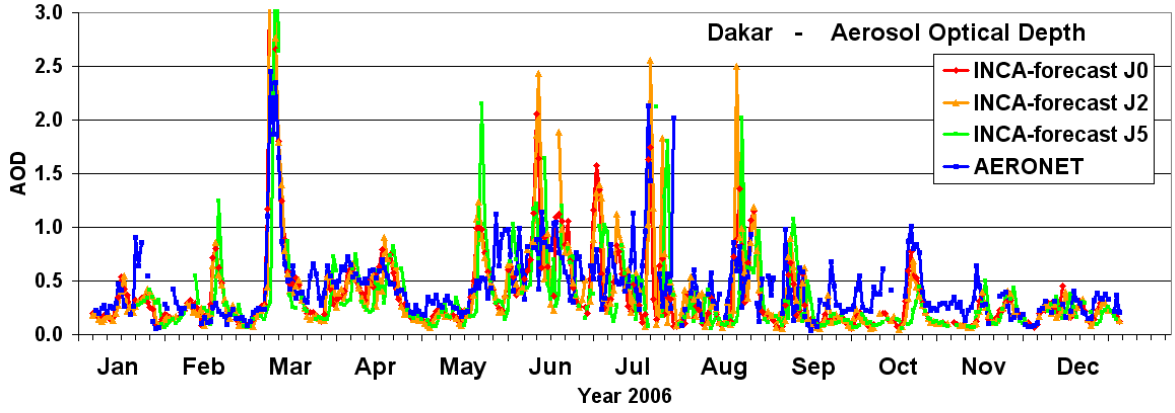

Figure 2 Mean daily total aerosol optical depth as measured at the AERONET/PHOTONS site near Dakar in West Africa by a sun photometer against the INCA model simulated values. Three modelled time series are shown: J0 corresponds to the day when the model is initialised, J2 corresponds to day two of the forecast and J5 uses the forecast five days ahead.

Other possibilities to judge the forecast model are hit rates and the error in predicting high levels of dust. Hit rates are established by calculating those occurrences when the model AOD level is within a range of 0.3 of

the observed value. Such hit rates attain 76%, 72% and 64% for the time steps J0, J2 and J5. This shows that the model can predict major dust events, which can result in high AOD values of well above 1 in the area as can be seen from figure 2. We also calculate the possible error in eventually issuing a warning for major dust events. We define here a major dust event as one with AOD values greater 0.5. Table 1 reports the cases when the model would have issued a correct or a false dust warning, in other words when it would have missed an event and when it correctly assumed that dust is below the warning level. For a threshold level of 0.5 AOD we find that at Dakar the model is correctly predicting on 71% (68%, 65%) of the days for time step J0 (J2, J5 respectively) the state of the dust load, be it over the threshold or below.

Table 1: Number of days in 2006, when the INCA model would be right/wrong with respect to predicting a dusty situation with levels above 0.5 AOD near Dakar. The error counts are based on the comparison of the daily Aeronet and INCA modelled AOD levels. Three time steps of the forecast model run and the reanalysis run are shown. Days with no information from Aeronet or from the forecast model are omitted.

#	J0	J2	J5	ReAnalysis
missing « warning »	43	47	52	40
correct « no warning »	118	116	114	124
correct « warning »	36	32	27	39
« warning » false	21	23	25	15

4.2. Example: Evaluation of the forward and assimilated IFS model versions EXLZ and EZUB with daily Aeronet aerosol optical thickness

Two model versions are now available from the GEMS-IFS system, which correspond a) to a forward run (EXLZ) as described by [*Morcrette et al.*, 2008] and b) to a reanalysis using an assimilation system based on satellite MODIS aerosol optical depth observations developed by [*Benedetti et al.*, 2008]. For the purpose of evaluation one year of 3-hourly simulated data have been downloaded and transformed to a daily data set of simulated aerosol optical depth (AOD). Corresponding aerosol optical depth from Aeronet have been compared and are documented via the AeroCom tools (http://nansen.ipsl.jussieu.fr/AEROCOM/data.html). The Aerocom tools now also allow for a visualisation of the daily AOD evolution, monthly maps of AOD and a simple evaluation statistics table from the comparison (see extract in table 2). As a reference we show the corresponding AeroCom Median model reflecting the average performance of other state-of-the-art global aerosol models.

It appears that the assimilated run outperforms the forward run in terms of correlation, RMS error, reproduction of the overall observed variability, the seasonal variability and the spatial variability. The assimilation procedure slightly increases the AOD in the model, probably due to a slight positive bias of MODIS over land, especially in high latitudes. Since the AeroCom median model is constructed from 10 other state of the art models it outperforms the individual models. The forward model version EXLZ performs as reported for other models and the comparison to the AeroCom Median model indicates that there is room for improvement for the aerosol module within the IFS model system. The assimilated run EZUB is at least as good as the reference AeroCom median model, though the years of reference are not the same. The inclusion of observational constraints in the assimilation procedure in form of the MODIS aerosol optical depth improves the simulation of fields of AOD. This can also be seen in figure 3 where the mean annual AOD fields are shown from MODIS, IFS runs EZUB and EXLZ.

Table 2: Comparison of two versions of IFS ECMWF model version for the year of 2003 with Aeronet aerosol optical depth (AOD) measurements at the Aeronet sites situated below 1000 m altitude. Data and model values are aggregated to monthly means from days with measurements (number of valid months used: #1280). The AeroCom Median model was constructed from 12 model simulations of the AeroCom A exercise (models as used in Textor et al., 2006). STD Mod/ STD Obs: Ratio of standard deviation in model and in observational data. Seasonal anomaly correlation is calculated after removing the mean concentration observed and modelled at every station from the monthly values. Spatial correlation reflects the correlation based on yearly means at all stations.

	Forward EXLZ 2003	Reanalysis EZUB 2003	AeroCom Median year 2000	Aeronet years 2003/2000
Mean AOD	0.24	0.27	0.16	0.22/0.18
Correlation	0.69	0.82	0.75	
RMS	0.13	0.11	0.10	
Std Mod / Std Obs	0.76	0.81	0.84	
Seasonal anomaly r	0.75	0.80	0.73	
Spatial r	0.71	0.78	0.66	

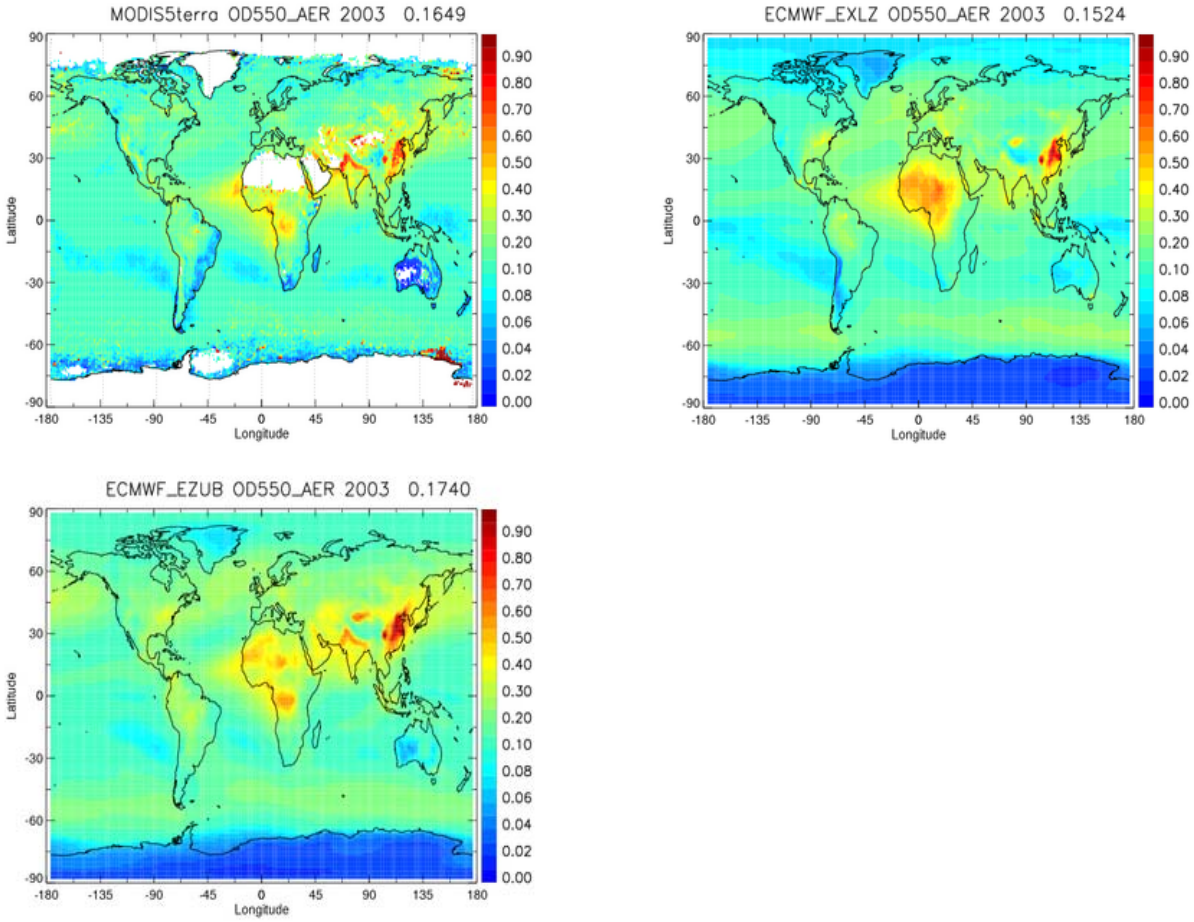

Table 3: Annual mean AOD@550nm for MODIS collection 5 from the Terra satellite, the IFS model forward run EXLZ and the IFS assimilated run EZUB for the year 2003.

The assimilated model run seems to better retrieve the spatial variability as for instance the higher AOD values in the Indian Ganges valley, in the position of the African biomass burning related aerosol plume, higher AOD values in California. Note that also for the global mean AOD, the assimilation results in a 15% higher value, as found at the Aeronet sites as documented in table 2.

One reason for the improved reproduction of the variability in the assimilated model run is the correction of the daily evolution of aerosol plumes. This can be shown on the basis of the evolution of the daily AOD at selected sites as shown in figure 4.

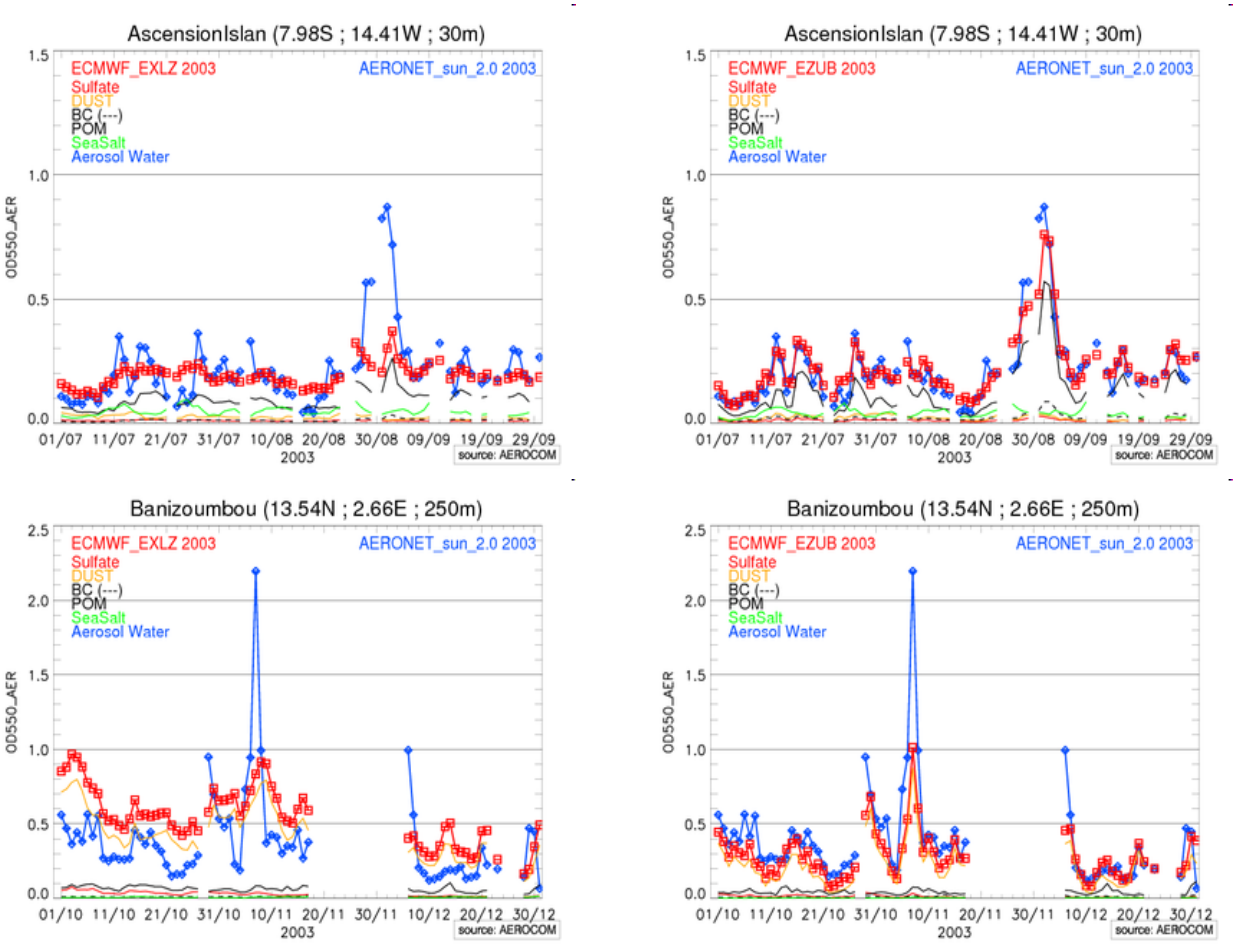

Table 4: Comparison example for daily evolution of AOD for two Aeronet sites. Left hand side is the forward simulations EXLZ, Right hand side the corresponding assimilation product using run EZUB.

The first example in figure 4 corresponds to a site in the southern Atlantic, influenced periodically by biomass burning plumes from Africa, especially in summer and autumn. The high variability from day to day reflects the changing transport conditions and periodic biomass burning. It is understandable that the resulting aerosol plumes, which travel over the ocean to Ascension island can be well observed by MODIS. The resulting correction during assimilation is thus very efficient and the simulated, assimilated AOD time series follows the Aeronet AOD closely. The second example concerns a dusty site in the Sahel zone. MODIS observations are less reliable over deserts and thus no AOD retrieval values are shown in figure 3 over the Sahara. However, it is clear from figure 4 that even for a Sahel site close to the Sahara area where no observations are available the 4D Var assimilation may improve the resultant AOD field considerable. Note that the Aeronet AOD values represent an independent evaluation of the assimilation result, since these observations have not been used in the assimilation.

5. Final Remarks

Further reducing the uncertainty in radiative forcing by aerosols is needed to significantly better predict and hindcast climate change. For this purpose it is important to significantly improve the evaluation process of regional and global aerosol models. Benchmark test tools currently being developed should be applied to

analyze model biases in more detail with respect to processes that govern concentrations and physico-chemical properties of the aerosol.

Several aerosol properties are responsible for uncertainty on the aerosol radiative forcing. Continued research is required to resolve the specific problems. An initial focus is on the vertical distribution of the aerosol and its impact on radiative forcing. One aspect of the vertical distribution – forcing relationship has become recently important through the analysis of the AeroCom results. Cloudy sky direct radiative forcing by aerosols may be responsible for a significant portion of model diversity for aerosol radiative forcing. The space borne lidar CALIOP offers for the first time the possibility to observe on larger scales aerosol above (and below) clouds and are now available for an entire year. These new observational data should be exploited in connection with new model simulations. A climatology of the relative position of the clouds compared to that of the aerosols needs to be established. It requires to be supplemented by sensitivity studies with individual models on the importance of the intervening factors of vertical mixing, injection height, absorbing properties and amplitude of emission strength of biomass burning. In general it is deemed necessary to improve the modelling of all relevant aerosol properties to be able to simulate second order aerosol effects such as aerosol-cloud interactions and other aerosol-climate feedbacks.

Aerosols are major carriers of matter from region to region. It might be difficult to reduce pollutant levels through local policy action if long-range transport brings in pollution from other regions. The HTAP task force has initiated perturbation experiments which are designed to study such long-range transport. They offer possibilities to explore from a mechanistic point of view differences among models. Perturbation experiments create regional tracers which can be followed independent of the global background. The destiny of these tracers should be more easily understood than that the background fields since the latter reflect the simultaneous action of very different regional climates. Tracer relationships should be employed to explore the sensitivity of the individual models to processes which regulate long distance transport such as convection or wet deposition.

Climate is expected to respond to aerosol perturbations in a short time. The Atmospheric Chemistry & Climate WCRP-SPARC/IGBP-IGAC initiative has proposed thus to activate a series of models to do a 20-25 year “hindcast” experiment to understand what has happened with tropospheric chemistry, and in particular ozone and aerosols in this period. Such an experiment allows to analyse observed changes in atmospheric composition over the past 20 years. Aerosol effects may be best validated for the recent past. Such studies must be then extended to the near future.

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