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## Modelling of strong heterogeneities in aerosol single scattering albedos over a polluted region

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[1] To date, most models dedicated to the investigation of aerosol direct or semi-direct radiative forcings have assumed the various aerosol components to be either completely externally mixed or homogeneously internally mixed. Some recent works have shown that a core-shell treatment of particles should be more realistic, leading to significant differences in the radiative impact as compared to only externally or well-internally mixed states. To account for these studies, an optical module, ORISAM-RAD, has been developed for computing aerosol radiative properties under the hypothesis of internally mixed particles with a  $n$ -layer spherical concentric structure. Mesoscale simulations using ORISAM-RAD, coupled with the 3D mesoscale model Meso-NH-C, have been performed for one selected day (06/24/2001) during the ESCOMPTE experiment in the Marseilles-Fos/Berre region, which illustrate the ability of this new module to reproduce spatial heterogeneities of measured single scattering albedo ( $\omega_o$ ), due to industrial and/or urban pollution plumes.

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### 1. Introduction

[2] It is now clearly recognized that anthropogenic aerosol particles have a climatic impact through three processes: directly, by scattering and/or absorbing the solar radiation [Haywood and Shine, 1995], indirectly, by modifying cloud properties [Twomey, 1977] and semi-directly, by heating atmospheric layers containing high concentrations of absorbing black carbon [Hansen *et al.*, 1997; Ackerman *et al.*, 2000].

[3] However, while the radiative forcing due to greenhouse gases may be determined to a reasonably high degree of accuracy, the uncertainties relating to aerosol radiative forcings remain large, due to the heterogeneous spatial and temporal distribution of aerosol chemical, physical and radiative properties. An improving on the level of confidence on the aerosol climatic effect requires a full range of in-situ or remote-sensing observations in selected regions as well as the development of regional and global models able to well simulate particle properties.

[4] The aim of this work is to present comparisons between measurements and modelling of aerosol radiative properties in the frame of a very documented experiment, ESCOMPTE [Cros *et al.*, 2004; Cachier *et al.*, 2005]. An

aerosol-optical module, ORISAM-RAD, has been developed to compute the radiative properties of internally mixed aerosols simulated by the microphysical-chemical ORISAM module included in the 3D mesoscale model Meso-NH-C [Bessagnet and Rosset, 2001; Cousin *et al.*, 2005; Liousse *et al.*, 2005]. In particular, light scattering and absorption are calculated using a  $n$ -layered spheres algorithm [Wu and Wang, 1991]. This is particularly pertinent when dealing with complex aerosol compositions in internal mixing state, involving black carbon, primary and secondary organics, inorganics and water.

[5] This new ORISAM-RAD module has been applied to the regional pollution episode of June 24th 2001. In this study, we focus our simulations on the aerosol single scattering albedo, which is the key aerosol optical property dominating the aerosol radiative effect [Hansen *et al.*, 1997] and which remains very difficult to estimate in all types of observation.

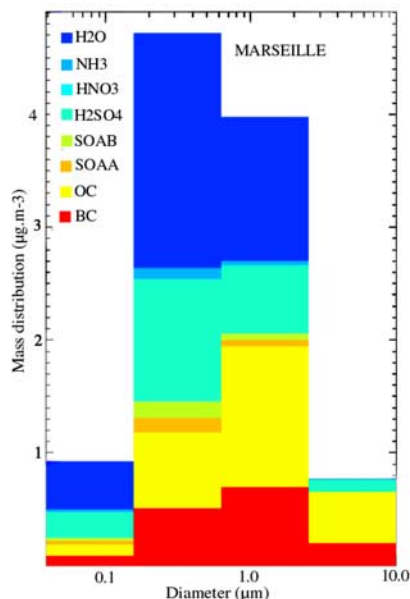
### 2. Aerosol Modelling

#### 2.1. Description of ORISAM Outputs

[6] Cousin *et al.* [2005] have simulated spatial and temporal distribution of aerosol size distributions and chemical properties over the ESCOMPTE domain for June 24th with the ORISAM (Organic and Inorganic Spectral Aerosol Module) module, which is implemented in the 3D mesoscale model Meso-NH-C (Mesoscale-Nonhydrostatic Chemistry model).

[7] In ORISAM, the representation of the aerosol size distribution is the sectional approach. The aerosol size distribution comprises four bins with diameters ranging from 0.03 to 10  $\mu\text{m}$  (Figure 1). Such a concept was retained for its flexibility in solving aerosol multicomponent systems involving the processes of nucleation, coagulation, condensation, adsorption, chemical reactions and deposition. Consequently, the aging of the sectional aerosol distribution may be followed.

[8] Six aerosol components are considered: primary carbonaceous aerosol, secondary organics, sulphates, nitrates, ammonium and water (Figure 1). Primary carbonaceous aerosol has been partitioned into black carbon (BC) and primary organic carbon ( $\text{OC}_p$ ) to account for their different physical and radiative properties. Secondary organics have been partitioned according to their origins, into an anthropogenic fraction (ASOA, Anthropogenic Secondary Organic Aerosol) and a biogenic one (BSOA, Biogenic Secondary Organic Aerosol). Secondary nitrates ( $\text{NO}_x$  oxidation in the gas phase and heterogeneous chemistry), ammonium (neutralization by nitric and sulphuric acids in the aerosol phase), sulphate and water are also considered in ORISAM.



**Figure 1.** Aerosol size distributions modelled by ORISAM (Organic and Inorganic Spectral Aerosol Module) at 12 UTC at Marseilles [Cousin *et al.*, 2005].

Primary organic matter ( $OM_p$ ) concentrations were calculated from the concentrations of primary organic carbon using a constant conversion factor [Lioussé *et al.*, 1996; Putaud *et al.*, 2000] and secondary organic components ( $OM_s$ ) were obtained by adding the concentrations of ASOA and BSOA.

## 2.2. Development of ORISAM-RAD

[9] The module we developed, ORISAM-RAD, computes the radiative properties of internally mixed aerosols simulated by the microphysical-chemical ORISAM module. In the module, the single particle extinction ( $Q_{ext}$ ), scattering ( $Q_{scat}$ ), absorption ( $Q_{abs}$ ) and the radiation pressure ( $Q_{pr}$ ) cross section efficiencies are computed using the n-layer spheres algorithm of Wu and Wang [1991], allowing us to model complex and detailed aerosol structure. This represents a great improvement as most of radiative forcing models generally assume aerosols as externally mixed [Tegen *et al.*, 1997; Takemura *et al.*, 2002] or internally homogeneously mixed [Haywood *et al.*, 1997; Myhre *et al.*, 1998]. The radiative properties are calculated for the total aerosol population using the methodology reported by Wu *et al.* [1996] for a sectional approach.

[10] In the case of our study (strongly polluted zone), each particle is assumed to be composed of three concentric different layers, where black carbon is assumed to be the

core. Primary organics are assumed to form the first shell around the black carbon core. The third layer is assumed to be composed by all water soluble hydrophilic components (ammonium sulphates, nitrates and  $OM_s$ ) in solution in water.

[11] One major advantage of ORISAM-RAD lies in the distinction between primary and secondary organic components. This allows, in this case, differentiated hygroscopic behaviours on their optical parameters, which has not been treated in regional or global radiative models until now. In our simulations, we assume primary components ( $OM_p$ ) as hydrophobic and secondary ( $OM_s$ ) as hydrophilic [Saxena *et al.*, 1995].

[12] The refractive indexes used in our simulations are reported in Table 1. The real and imaginary parts of the third layer have been determined using a volume average procedure [Lesins *et al.*, 2002] for all water soluble components. The aerosol refractive indexes of hydrophilic aerosols have been computed at the ambient relative humidity using the methodology described by Mallet *et al.* [2003].

[13] One critical point in radiative computations concerns the choice of the black carbon refractive index. Sensitivity tests have been performed indicating a small error on the aerosol single scattering albedo of  $\pm 0.02$ . Similar tests have been made to estimate the error on  $\omega_o$  due to the uncertainty on the black carbon aerosol density ( $1000 < \rho_{bc} < 1500 \text{ kg.m}^{-3}$ ) indicating an error of  $\pm 0.02$ .

## 3. In-Situ $\omega_o$ Observations

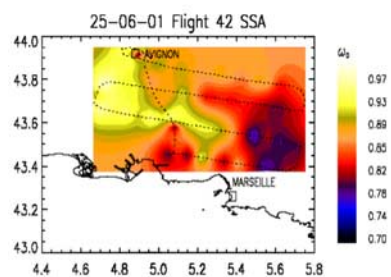
[14]  $\omega_o$  was not directly measured and preparations of experimental data have been performed. Firstly, nephelometer and aethalometer measurements performed on-board the ARAT aircraft [Mallet *et al.*, 2005] have been used to obtain a regional picture of  $\omega_o$  over Marseilles-Fos/Berre area (Figure 2). It should be noted that we used values obtained for June 25th, as no values are available for the 24 June. However, the meteorological conditions have been shown to be quite similar for these two days [Cros *et al.*, 2004].

[15] In a second time, we used chemical and optical measurements performed by Mallet *et al.* [2003] to obtain a  $\omega_o$  reference value at surface ( $\omega_o \sim 0.75$  at 550 nm) in a peri-urban site near Marseilles. Finally, we used AERONET observations for June 24th and 25th, at the Avignon and Vinon sites, located in the industrial plume, using the

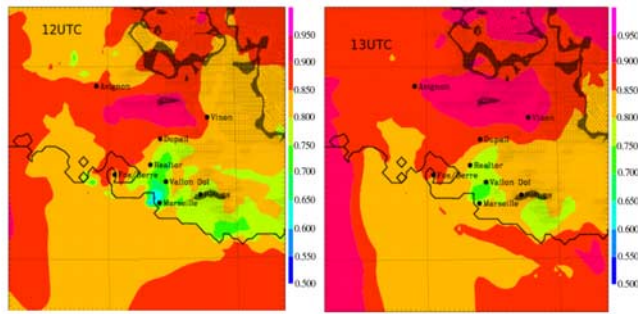
**Table 1.** Dry Aerosol Refractive Indexes (at 550 nm) Used in Optical Simulations<sup>a</sup>

Aerosol Species	n	k	References
BC	1.87	0.569	Marley <i>et al.</i> [2001]
OM	1.45	$1.00 \times 10^{-3}$	Krekov [1993]
Ammonium sulphate	1.53	$1.00 \times 10^{-7}$	Toon <i>et al.</i> [1976]
Nitrate	1.53	$6.00 \times 10^{-3}$	d'Almeida <i>et al.</i> [1991]
Liquid water	1.34	$1.80 \times 10^{-8}$	Hale and Query [1973]

<sup>a</sup>Here n is the real and k the imaginary parts of refractive indexes.



**Figure 2.** Aerosol single scattering albedos estimated at 550 nm from aircraft measurements June 25, 2001 at 12 UTC.



**Figure 3.** Aerosol single scattering albedos (at 550 nm) simulated at surface for June 24, 2001 during ESCOMPTE at 12 UTC and 13 UTC.

Dubovik and King's [2000] inversion algorithm in order to obtain  $\omega_o$  reference values in the atmospheric column.

#### 4. Regional Simulations and Results

[16] The modeling  $\omega_o$  values obtained by our simulations have been compared with the ones measured during ESCOMPTE. Simulation of spatial distribution of surface aerosol single scattering albedos (at 550 nm) for June 24th at 12 UTC and 13 UTC are displayed in Figure 3. Figure 3 clearly shows strong  $\omega_o$  heterogeneities in the domain, with two aerosol plumes characterized by different single scattering albedos. The first plume, issued from the urban area of Marseilles is characterized by surface single scattering albedos around 0.70, denoting strong absorbing aerosols. This modelling result is well consistent with the value ( $\sim 0.75$ ) obtained at ground level (see previous paragraph, Mallet *et al.* [2003]). This result is also consistent with the chemical composition of particles collected at Marseilles, with sub-micronic aerosols containing about 11% of black carbon [Cachier *et al.*, 2005].

[17] The second aerosol plume, issued from the industrial area of Fos/Berre, is characterized by single scattering albedos about 0.92 at 550nm, revealing weak absorbing aerosols. This is also consistent with the chemical measurements performed at the Martigues industrial site, with sub-micronic particles containing about 4% BC [Cachier *et al.*, 2005]. We observe quite a good agreement between  $\omega_o$  simulated in the plume and the ones retrieved from the AERONET network at Avignon and Vinon ( $0.94 \pm 0.03$  and  $0.92 \pm 0.03$ , respectively), both sites located within the Fos/Berre plume.

[18] We thus can observe a good reproduction by ORISAM-RAD of the  $\omega_o$  geographical pattern and magnitude obtained over the domain from aircraft in-situ measurements. Indeed, two aerosol plumes issued from Marseilles ( $\omega_o \sim 0.75$  at 550 nm) and Fos/Berre ( $\omega_o \sim 0.95$ ), observed in Figure 2 are well simulated by ORISAM-RAD. Such  $\omega_o$  heterogeneities in the domain result also in large heterogeneities of the direct and semi-direct radiative impacts at regional scale. Though the two anthropogenic plumes reduce the solar radiation reaching the surface, their radiative effects into the PBL and at the top of atmosphere are antagonistic. The first (urban) plume absorbs short wavelength of the solar radiation in the PBL, unlike the second

one (industrial area of Fos/Berre) reflects radiations back to space, respectively heating and cooling the atmosphere.

#### 5. Conclusion

[19] Our study introduces a coupled aerosol-radiative module, ORISAM-RAD, with the 3D mesoscale model Meso-NH-C, for particulate radiative properties and climatic impact estimation. One main advantage is to treat aerosol particles under the internal mixing hypothesis based upon a n-layer spheres algorithm. The ORISAM-RAD module has been applied to a pollution episode during the ESCOMPTE 2001 campaign. Regional simulations have displayed high spatial  $\omega_o$  heterogeneities, in close connection with the geographical patterns observed from aircraft measurements, revealing two anthropogenic aerosol plumes, one industrial and the other urban, characterized by quite different surface single scattering albedos, respectively about 0.94 and 0.70 at 550 nm. We are now implementing a radiative transfer model using the optical properties simulated by ORISAM-RAD in order to investigate the differentiated radiative forcing due to anthropogenic particles and its potential feedbacks on regional dynamics. This flexible ORISAM-RAD module will be also implementing in regional and global models for climate studies.

[20] **Acknowledgments.** The authors are indebted to Dr. Bernard Cros, Dr. Pierre Durand and Dr. H. Cachier for the organization of the ESCOMPTE experiment. We gratefully thank the different Institutes in France (CNRS, INSU, ADEME, Ministry of Environment) for their support. We would like also to thank F. Cousin, B. Guillaume and R. Rosset for their help in this work.

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