Impact of Aerosol Microphysical Properties on Mass Scattering Cross Sections

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Abstract—***We assessed the sensitivity of simulated mass scattering cross sections (α_λ^{sca} $[m^2/g]$) of three aerosol species to perturbed particle microphysical properties and derived constraints on these microphysical properties, suitable for the north-western Mediterranean basin, from a comparison between code calculations and observations. In detail, we calculated α_λ^{sca} of mineral dust, organic carbon and sulfate at three wavelengths in the visible range with a T-matrix optical code, considering $\pm 20\%$ perturbations on size distribution, refractive index and mass density, and spheroids with two different axial ratios as shape perturbations. Then, we compared the simulation results with a set of observed α_λ^{sca} of mineral dust, aged organics and ammonium sulfate sources provided by the Institute of Environmental Assessment and Water Research (IDAEA-CSIC) and representative of the north-western Mediterranean basin.***

I. Introduction

Atmospheric aerosols can scatter and absorb electromagnetic radiation, causing a redistribution of the radiative energy in the atmosphere [1]. The Aerosol-Radiation Interaction (ARI) radiative forcing still contributes to dominate the uncertainty associated with the anthropogenic contribution to the climate change [2]. Also the role of the natural aerosols in affecting the Earth's radiative balance through ARI is poorly constrained [3]. The ARI parameterization mainly consists in the characterization of the aerosol optical properties, which in turn depend on the microphysical properties of the particles [1], [4]. Hence, errors in the particle microphysical assumptions can affect the calculation of the optical properties and so the assessments of the ARI radiative effects [5], [6]. For this reason, we consider a study on the relationship between microphysical and optical properties a recommendable first step to better parameterize the ARI in the atmospheric models.

II. T-matrix code

The T-matrix method is a powerful exact technique for computing light scattering by non-spherical particles [7]. For this work, the T-matrix code by [7] has been used. It allows calculating integrated optical properties of poly-disperse, randomly oriented, rotationally symmetric, homogeneous particles.

III. Observations

We used a set of observed aerosol α_λ^{sca} provided by the IDAEA-CSIC. [8] collected measurements of dry ($RH < 40\%$) aerosol PM_{10} mass concentrations (gravimetric masses from 24h filters) and optical properties (scattering coefficients with Nephelometer AURORA 3000, Ecotech), in the period 2010-2014, at the Montseny regional background station (middle altitude emplacement within the Montseny Natural Park, Spain: $41°46'45,63"N$ - $02°21'28,92"E$; $720m$ a.s.l.). From these data, [9] derived α_λ^{sca} at 3 visible wavelengths ($\lambda_1 = 0,450\mu m; \lambda_2 = 0,525\mu m; \lambda_3 = 0,635\mu m$) through Multilinear Regression (MLR) analysis applied to aerosol sources [10] detected in the PM_{10} mass chemical speciated data [11] through the application of the Positive Matrix Factorization (PMF) model [12]. In our work we used three of the seven sources detected at the Montseny station: mineral dust (typical crustal elements), aged organics (mainly organic carbon from biogenic sources but with a significant contribution from biomass burning) and ammonium sulfate (mainly SO_4^{2-} and NH_4^+) [10]. These data can be considered representative of the regional background of the north-western Mediterranean basin.

IV. Experiment setup

We defined the reference microphysical properties following the aerosol parameterization used in the NMMB-MONARCH model (formerly known as NMMB/BSC-CTM) [13], [14], [15], which in turn is based on the OPAC database [16], but with some deviations. We performed calculations of integrated α_λ^{sca} of mineral dust, organic carbon and sulfate, The calculations have been performed at the same three visible wavelengths of the observations ($\lambda_1 = 0,450\mu m; \lambda_2 = 0,525\mu m; \lambda_3 = 0,635\mu m$). Then, in order to generate variability in the microphysical assumptions for the particles, we gave as inputs to the T-Matrix code the reference microphysical properties independently perturbed. In particular, for each aerosol species, we considered perturbations of $\pm 20\%$ on size distribution (separately for geometric radius r_g and standard deviation σ_g), refractive index (separately for real n_R and imaginary n_I parts) and mass density ρ; as perturbations for the spherical shape we considered two types of spheroid: moderate and extreme, averaging for each axial ratio χ the oblate and the prolate options.

V. Data analysis

We compared simulated and observed α_λ^{sca} associating mineral dust with the mineral dust source, organic carbon with the aged organics source and sulfate with the ammonium sulfate source. At first, we evaluated the sensitivity of α_λ^{sca} values and spectral dependence to the different perturbed microphysical properties. Regarding the α_λ^{sca} values, for each species and microphysical property we estimated the extent of the variability range (generated by the perturbations) of the simulated value at $\lambda_2 = 0,525\mu m$ respect to the uncertainty affecting the observed value at the same wavelength. Regarding the α_λ^{sca} spectral dependence, instead, we estimated the extent of the variability range of the log-linear spectral dependence slope of the simulated values respect to the uncertainty affecting the slope of the observed values. The spectral dependence slopes have been derived by performing linear fits of the logarithmic spectral dependence of both simulated and observed α_λ^{sca}, taking into account

Table I
PRESCRIPTIONS DERIVED FROM OUR ANALYSIS FOR SIZE DISTRIBUTION (SD), REFRACTIVE INDEX (RI), MASS DENSITY (DN) AND SHAPE (SH) OF MINERAL DUST (DU), ORGANIC CARBON (OC) AND SULFATE (SU). THE VALUES REPORTED FOR THE MICROPHYSICAL PARAMETERS MARKED BY * ARE REFERENCE VALUES, FOUND TO BE SUITABLE FOR THE CORRESPONDENT SPECIES IN THE GEOGRAPHYICAL AREA UNDER STUDY.

		DU	OC	SU
SD:	$r_g(\mu m)$	$3,583 \cdot 10^{-1}$	$2,544 \cdot 10^{-2}$	$8,340 \cdot 10^{-2}$
	σ_g	$1,600$	$1,760$	$1,624$
	$r_{eff}(\mu m)$	$6,221 \cdot 10^{-1}$	$5,656 \cdot 10^{-2}$	$1,501 \cdot 10^{-1}$
RI:	n_R	$(1,530; 1,530; 1,530)(^*)$	$(1,576; 1,576; 1,576)$	$(1,547; 1,545; 1,543)$
	n_I^*	$(8,500; 6,650; 4,500) \cdot 10^{-3}$	$(1,730; 1,250; 0,696) \cdot 10^{-2}$	$(1,000; 1,000; 1,610) \cdot 10^{-8}$
DN:	$\rho^*(kg/m^3)$	$2,506 \cdot 10^3$	$1,800 \cdot 10^3$	$1,700 \cdot 10^3$
SH:	χ^*	$1,000$	$1,000$	$= 1,000$

the measurement errors as weights for the observation fits. In order to complete the comparison analysis and to constrain the perturbed microphysical properties, then, we performed a compatibility test on the best fit parameters. The test has been performed, for each aerosol species, on the best fit parameters of all the perturbed simulations respect to those of the correspondent observations.

VI. RESULTS

We found that the mineral dust α_λ^{sca} values are only affected by the size distribution and, with a lower impact, by the mass density perturbations. On the other hand, no microphysical properties seem to have any impact on the α_λ^{sca} spectral dependence. So, it appears that, due mainly to the size of the particles bigger than the visible wavelengths, the dust α_λ^{sca} are quite stable respect to the microphysical perturbations (in the spectral range of the experiment). The organic carbon α_λ^{sca} values are affected mainly by refractive index (real part), size distribution and, with a lower impact, by mass density and shape perturbations. The α_λ^{sca} spectral dependence, instead, is significantly affected only by the size distribution perturbations. So, the organic carbon α_λ^{sca} appear less stable than the mineral dust ones respect to the microphysical perturbations, due mainly to the size of the particles smaller than the visible wavelengths. The sulfate α_λ^{sca} values are mainly affected by the refractive index (real part) and, with a lower impact, by size distribution, mass density and shape perturbations. On the other hand, the α_λ^{sca} spectral dependence is only affected by size distribution and refractive index perturbations. So, it seems that the sulfate α_λ^{sca} are the most unstable respect to the microphysical perturbations, due to the particle size approximately comparable with the visible wavelengths. The microphysical prescriptions derived from our analysis for the three aerosol species, suitable for the north-western Mediterranean Basin, are reported in Table I.

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