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Sensitivity of high-spectral resolution and broadband thermal infrared nadir instruments to the chemical and microphysical properties of secondary sulfate aerosols in the upper-troposphere/lower-stratosphere

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The observation of upper-tropospheric/lower-stratospheric (UTLS) secondary sulfate aerosols (SSA) and their chemical and microphysical properties from satellite nadir observations (with better spatial resolution than limb observations) is a fundamental tool to better understand their formation and evolution processes and then to estimate their impact on UTLS chemistry, and on regional and global radiative balance. Thermal infrared (TIR) observations are sensitive to the chemical composition of the aerosols due to the strong spectral variations of the imaginary part of the refractive index in this band and, correspondingly, of the absorption, as a function of the composition Then, these observations are, in principle, well adapted to detect and characterize UTLS SSA. Unfortunately, the exploitation of nadir TIR observations for sulfate aerosol layer monitoring is today very limited.

Here we present a study aimed at the evaluation of the sensitivity of TIR satellite nadir observations to the chemical composition and the size distribution of idealised UTLS SSA layers. The sulfate aerosol particles are assumed as binary systems of sulfuric acid/water solution droplets, with varying sulphuric acid mixing ratios. The extinction properties of the SSA, for different sulfuric acid mixing ratios and temperatures, are systematically analysed. The extinction coefficients are derived by means of a Mie code, using refractive indices taken from the GEISA (Gestion et Étude des Informations Spectroscopiques Atmosphériques: Management and Study of Spectroscopic Information) spectroscopic database and log-normal size distributions with different effective radii and number concentrations. High-spectral resolution pseudo-observations are generated using forward radiative transfer calculations performed with the 4A (Automatized Atmospheric Absorption Atlas) radiative transfer model, to estimate the impact of the extinction of idealised aerosol layers, at typical UTLS conditions, on the brightness temperature (BT) spectra observed by satellite instruments. We isolated a marked and typical spectral signature of these aerosol layers between 700 and 1200 cm^{-1}, due to the absorption bands of the sulfate and bisulfate ions and the undissociated sulfuric acid, with the main absorption peaks at 1170 and 905 cm^{-1} (sulfuric acid vibrational bands).

The dependence of the residual aerosol spectral BT signature to the sulfuric acid mixing ratio, and effective number concentration and radius, as well as the role of interfering parameters like the ozone, sulfur dioxide, carbon dioxide and ash absorption, and temperature and water vapour profile uncertainties, are analysed and critically discussed. The information content (degrees of freedom and retrieval uncertainties) of synthetic satellite observations is estimated for different instrumental configurations. High spectral resolution (Infrared Atmospheric Sounding Interferometer (IASI)-like pseudo-observations) and broadband spectral features (Moderate Resolution Imaging Spectroradiometer (MODIS) and Spinning Enhanced Visible and InfraRed Imager (SEVIRI)-like pseudo-observations) approaches are proposed and discussed.