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Polarized radiative transfer through terrestrial atmosphere accounting for rotational Raman scattering

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ABSTRACT

This paper is devoted to the phenomenological derivation of the vector radiative transfer equation (VRTE) accounting for first-order source terms of rotational Raman scattering (RRS), which is responsible for the in-filling of Fraunhofer and telluric lines by inelastic scattered photons. The implementation of the solution of the VRTE within the framework of the forward-adjoint method is given. For the Ca II and the oxygen A-band (O_2 A) spectral windows, values of reflectance, degree of linear polarization (DOLP) and in-filling, in zenith and nadir geometry, are compared with results given in literature. Moreover, the dependence of these quantities on the columnar loading and vertical layering of non-spherical dust aerosols is investigated, together with their changes as function of two habits of ice crystals, modeled as regular icosahedra and severely rough aggregated columns. Bi-directional effects of an underlying polarizing surface are accounted for. The forward simulations are performed for one selected wavelength in the continuum and one in the strong absorption of the O_2 A, as their combination can be exploited for the spaceborne retrieval of aerosol and cloud properties. For this reason, we also mimic seasonal maps of reflectance, DOLP and in-filling, that are prototypical measurements of the Ultraviolet-Visible-Near Infrared (UVN) sensor, at a nominal spectral resolution of 0.12 nm. UVN is the core payload of the upcoming European Sentinel-4 mission, that will observe Europe in geostationary orbit for air quality monitoring purposes. In general, in the core of O_2 A, depending on the optical thickness and altitude of the scatterers, we find RRS-induced in-filling values ranging from 1.3% to 1.8%, while DOLP decreases by 1%. Conversely, while negligible differences of RRS in-filling are calculated with different ice crystal habits, the severely rough aggregated column model can reduce DOLP by a factor up to 10%. The UVN maps of in-filling show values varying between 1% and 8%. These changes are mainly driven by surface type and seasonal observational geometry. However, accounting for RRS, differences in DOLP do not exceed ± 0.2% within the full instrumental field-of-view.

1. Introduction

The filling of Fraunhofer lines in skylight scattered by air molecules was first reported by Shefov [1] and independently discovered by Grainger and Ring [2] a few years later. This phenomenon, labeled as the Ring effect (after one of its discoverers) and its origin has been subject of numerous theoretical and observational studies. One pioneering measurement study was the one of [3]. They repeated the observations by Grainger and Ring [2] and found that the light inside the Fraunhofer lines has lower degree of polarization than in the surrounding spectral continuum. Subsequent investigations by several research teams were also considering unpolarized light and attributed the effect to mainly one physical process: rotational Raman scattering at air molecules. For a comprehensive literature review related to this conclusion the reader is referred to [4] and to later updates from [5, among others].

Following the study of [3], measurements of skylight polarization have been performed starting with [6] (in the UV), [7] (in the Na-D2) and [8] (Hβ) showing that the Degree Of Linear Polarization (DOLP) in Fraunhofer lines is lower than in the continuum, hence confirming the assumption that RRS is the predominant source of in-filling. Using polarization measurements and based on the assumption that RRS depolarizes light, Solomon et al. [9] introduced an empirical method to determine effective Ring effect cross-sections, which are, to date, used in Differential Optical Absorption Spectroscopy (DOAS) applications.

A series of measurements by Clarke and Basurah [10], Basurah [11,12] and Clarke and Naghizadeh-Khouei [13] reported events when DOLP in the center of the Fraunhofer lines was significantly higher than that in the continuum. However, this finding is in

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contradiction to the contemporary understanding of the processes in charge (Rayleigh, Cabannes and rotational Raman scattering). On the other hand, [14], while performing similar measurements as the ones mentioned above, report “exclusively lower polarization in the Fraunhofer lines in accordance with rotational Raman scattering”.

To be able to analyze conforming or non-conforming behavior of the measurements with respect to our model-based understanding, polarization must be accomodated into the radiative transfer codes prepared to account for RRS. While the first extensive radiative transfer study incorporating polarization and RRS for ground-based observations has been published by Humphreys et al. [15], only several years later [14] and [16] resumed these efforts and modeled polarized radiation up to the second order of scattering taking into account RRS. The agreement with measurements based mainly on work in the UV between the simulated and the observed polarization spectra was considered to be good. Landgraf et al. [17] extended the approach, accounting for polarization, to higher orders of scattering and confined themselves to first-order-RRS. For the sake of minimization of the computational burden, van Deelen et al. [18] simplified the modeled atmosphere and introduced again the unpolarized case but were able to show the insignificance of higher-order RRS in the UV at moderate spectral resolution.

While mentioned literature was often confined to the UV, following studies looked at the near-infrared (NIR) part of the spectrum and, in particular, at the strongest band of molecular oxygen centred about 761 nm (the O_2A-band). For instance, Pfeilsticker et al. [19] speculated that RRS should not play an important role in the in-filling of the A-band, due to the dominating effect of Mie scattering by aerosols and clouds over molecular scattering. However, their measurements were carried out in zenith geometry only beyond 763 nm, for which the individual lines of the A-band were not appreciably filled, as reported by Sioris and Evans [20]. Conversely, in satellite nadir geometry, Sioris and Evans [20] acknowledged the impact of the Ring effect and concluded describing the interplay between elastic Mie scattering and inelastic RRS in the in-filling of the band. They also looked at the dependence of Ring in-filling on viewing geometry, and briefly on ground albedo, aerosol, and cloud properties. Only later, Vasilkov et al. [21] provided additional results and, similar to the conclusions drawn by de Beek et al. [22] in the UV, found the consistent tendency of a decreasing Ring effect as a result of intervening atmospheric scattering layers. While these studies took into account RRS and neglected polarization in the O_2A-band, other research dealt with polarization effects, although neglecting RRS, in the presence of water clouds [23], cirrus clouds [24] and aerosols [25]. The results of the last two studies are of special interest because the simulations were performed at the rather high spectral resolution of 0.044 nm, mimicking measurements of the Orbiting Carbon Observatory (OCO) mission [26]. Still, none of the cited research prescribed in the calculations a directional surface reflectivity and limited their results to a underlying Lambertian surface.

Having all this in mind, we present the assessment of the magnitude of the in-filling and polarization together not only for Fraunhofer lines in the UV but also in the O_2A-band for a clear atmosphere and with the inclusion of dust-like aerosols and ice clouds. This research is timely and becoming increasingly important because of the advancements in instrument design. Spectrometers of increasing spectral resolving power are used and often they are sensitive to polarization, even in the case that they do not directly measure it. For this reason, we also study Ring and polarization for fictitious measurements of the Ultraviolet-Visible-Near infrared (UVN) instrument [27] onboard the upcoming Meteosat Third Generation (MTG-S) platform, managed by EUMETSAT, as core payload. This is referred to as the Sentinel-4 mission and is part of the European spaceborne air quality monitoring programme Copernicus [28]. UVN builds on the heritage of the Global Ozone Monitoring Experiment (GOME, [29]) and the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY, [30]) for the high spectral resolution in the UV–vis-NIR range, on the Ozone Monitoring Instrument (OMI, [31]) for the high spatial resolution and on the GeoSCIA concept [32] for its geostationary orbit. Hence, UVN is equipped with a two-dimensional (2-D) charge-coupled device (CCD) camera and, with a push-broom technique, aims at hourly measurements of atmospheric and surface constituents such as trace gases, aerosols and clouds mainly above Europe and neighbouring regions. Clouds and aerosols, in turn, are a key input for the accurate retrieval of gaseous tropospheric species, which will be retrieved from the UV–vis at a resolution of 0.5 nm. It has been already shown that, in this case, the RRS spectral signature is considerably flattened even at strongly absorbing wavelengths [21], so it is reasonable to neglect Ring effect in this spectral range. However, the algorithms selected to operationally retrieve cloud and aerosol properties, such as height and optical thickness [33,34], will make use of the oxygen A-band at a resolution of 0.12 nm, for which the in-filling due to inelastic scattering is not negligible anymore [21]. Moreover, considering the UVN footprint of 8×8 km^2 together with the hourly sampling and the relative evolution of observational geometry, it becomes also clear that the directionality of surface reflectivity must be taken into account.

To test the assumption that RRS plays the predominant role in the filling of Fraunhofer (and telluric gas absorption) lines and to compensate for its impact on trace gas retrievals in the UV/Vis spectral ranges, several research groups have developed and afterward updated radiative transfer models (RTM) accounting for RRS [4,5,14–18,35–42].

Our work has the purpose to continue the achieved progress in the field of radiative transfer modeling by updating the RTM SCIATRAN [43]. SCIATRAN is a comprehensive software package aiming the modeling of radiative transfer processes in the terrestrial atmosphere and ocean in the spectral range from the ultraviolet to the thermal infrared (0.18-40 µm) including multiple scattering processes, polarization, thermal emission, bi-directional reflectance distribution function (BRDF) effects or, alternatively, ocean-atmosphere coupling. While the main focus of the recent paper by Rozanov et al. [44] is the exploitation of vibrational Raman scattering from the scalar in-filling of Fraunhofer lines for the retrieval of broadband fluorescence induced by chlorophyll or dissolved organic matter in water, this paper extends the framework of the RTM explicitly including RRS to polarization. This task is accomplished with the aid of the forward-adjoint technique instead of the standard discrete ordinates method, as in [44]. It builds-up upon the work published in [43] and [42] and focuses on atmospheric scatterers of specific optical properties, supporting the interpretation of real measurements of the oxygen A-band by Sentinel-4.

This paper starts with the mathematical derivation of the radiative transfer equation accounting for RRS and polarization (Section 2) and then proceeds to the comparison (Section 3) of our results with previously published ones. Sections 3.1 and 3.2 deal with the comparisons of results in the Fraunhofer lines and Section 3.3 in the O_2A-band. The surface and atmospheric setup needed for the RT calculations is described in Section 4.1. Results in and outside the O_2A-band are given in Section 4.2 for a clear and a dust-laden scenario. Results for ice clouds are given in Section 4.3, while seasonal maps of reflectance, Ring in-filling and DOLP for the UVN geometry are portrayed in Section 4.4. In Section 5 the main results are highlighted and conclusions are drawn. Lastly, the derivation of the diffuse component of the radiation field and its numerical implementation as Fourier series are given in Appendix A and Appendix B.