3.8 The NASA algorithm

The National Aeronautics and Space Administration (NASA) Real Time algorithm is developed as a quick look algorithm that relies on the fact that atmospheric scattering strongly affects DOAS measured O_4 absorption (Spinei et al., Fast aerosol extinction coefficient profile estimation from MAXDOAS UV/VIS measurements, in preparation). Two separate approaches are used for aerosol and trace gas profile retrieval. The aerosol profile algorithm determines the layer aerosol extinction coefficients by comparing measured Ring and O_4 absorption with Ring and O_4 absorption under pure Rayleigh conditions. Air mass factors and Ring absorption for the Rayleigh case are pre-calculated using the VLIDORT v2.8 and LIDORT-LRRS v2.5 radiative transfer models, respectively (Spurr et al., 2008; Spurr, 2008) assuming the U.S. standard atmosphere. Since Ring simulations were not provided in this study, aerosol analysis was performed only at 477 nm. O_4 dSCDs are corrected for SZA dependence. Eq. 10 is the simplified equation used in this study to calculate aerosol scattering extinction coefficients at each layer for specific observation geometry (EA and RAA) Θ. We also assume an aerosol single scattering albedo of $\omega_{aer}(\lambda) = 1$.

$$\epsilon_{aer}(\lambda, \Theta, \vartheta) \approx \frac{\tau_{\mathrm{O4}}^{\mathrm{noaer}} - \tau_{\mathrm{O4}}^{\mathrm{aer}}}{\Delta h} \tag{10}$$

with $\tau_{\mathrm{O4}}^{\mathrm{aer}}$ and $\tau_{\mathrm{O4}}^{\mathrm{noaer}}$ being the optical density with and without aerosols in the respective layer, λ denoting wavelength, and ϑ the SZA. The thickness Δh of the respective layer is determined from the corrected O_4 dSCD using simple trigonometry according to Eq. 11 and 12, resulting in an atmosphere specific grid:

$$h_i = \frac{\Delta S_{\mathrm{O4}}(\alpha_i) + V_{\mathrm{O4}}}{n_{\mathrm{O4}}} \cdot \sin \alpha_i \tag{11}$$

$$\Delta h = h_{i+1} - h_i \tag{12}$$

with $S_{\mathrm{O4}}(\alpha_i)$ being the O_4 slant column density at elevation angle α_i and V_{O4} the O_4 vertical column density.

The maximum number of vertical layers is equal to the number of elevation angles. Profiles are considered invalid if less than four measurements are used in the profile calculation, with all of the synthetic data analysed here satisfying this test. Within this study, an exponential profile reducing to 0.01% of the last altitude layer extinction coefficient at 4 km was added for consistency with the other algorithms. The resulting profile was then linearly interpolated on the common grid (200 m up to 4 km).

The trace gas profile retrieval does not rely on the aerosol retrieval. The trace gas VCD V_{gas} is calculated first from the trace gas and O_4 dSCD measurements at 15° EA, $\Delta S_{\mathrm{gas}}^{15^\circ - 90^\circ}$:

$$V_{\mathrm{gas}} = \frac{\Delta S_{\mathrm{gas}}^{15^\circ - 90^\circ}}{\Delta A_{\mathrm{O4}}^{15^\circ - 90^\circ}} \tag{13}$$

with the according O_4 dAMF $\Delta A_{\mathrm{gas}}^{15^\circ-90^\circ}$ calculated via

$$A_{\mathrm{O4}}^{15^\circ-90^\circ} = \frac{\Delta S_{\mathrm{O4}}^{15^\circ-90^\circ}}{V_{\mathrm{O4}}} + 1 \tag{14}$$

Near-surface trace gas VMR M_{gas} are calculated by simple extrapolation of trace gas and O_4 dSCDs at 1° and 2° EA to the surface, yielding $\Delta S(\alpha,\lambda)_{\mathrm{gas}}^{\mathrm{extrapolated}}$ and $\Delta S(\alpha,\lambda)_{\mathrm{O4}}^{\mathrm{extrapolated}}$, and by converting to the VMR using surface pressure p and temperature T similar to Sinreich et al. (2013):

$$M_{\mathrm{gas}} = \frac{\Delta S(\alpha,\lambda)_{\mathrm{gas}}^{\mathrm{extrapolated}}}{\Delta S(\alpha,\lambda)_{\mathrm{O4}}^{\mathrm{extrapolated}}} \cdot \frac{p \cdot N_a}{R \cdot T} \cdot \chi_{\mathrm{O2}}^2 \tag{15}$$

The rest of the profile VMR is calculated using O_4 and trace gas dSCDs at multiple EAs. The layer altitude is calculated similar to the aerosol case. The derived profile is then converted to partial columns and scaled by the total VCD. As in the aerosol case, the layer grid is condition specific and was adjusted to the common grid in this study.

4 Model scenarios and RTM settings

A first important step for the comparison of retrieval algorithms is the assessment of their capability to realistically simulate the underlying physical processes using appropriate forward models, which are in this case atmospheric radiative transfer models. This section describes the forward model parameters and atmospheric scenarios for the modelling of O_4, NO_2 and HCHO SCDs, while the comparison of forward modelled SCDs is presented in Section 5.

The model atmosphere for the forward calculations consists of 67 layers, with a resolution of 100 m at altitudes between the surface and 4 km, and a coarser resolution above. Note that the retrieval of extinction and trace gas profiles is performed on a coarser grid with 200 m resolution in the lowermost 4 km. The choice of a finer grid for the forward modelling than for the inverse modelling allows for the investigation of the impact of sub-grid trace gas and aerosol variabilities on the retrieved profiles. For the forward modelling, a constant concentration within each layer has been implemented for the model calculations whenever possible.

The trace gas concentration and aerosol extinction profile scenarios for the forward modelling of HCHO, NO_2 and O_4 SCDs are shown in Figure 2. The same set of trace gas profiles is assumed for both HCHO and NO_2, in accordance with ambient measurements in mid-latitudes, where typical concentrations of both species are of the same order of magnitude (e.g., Vlemmix et al., 2015b). For simplicity, it is assumed that aerosol extinction does not change with wavelength (Ångström exponent of zero). The model profiles are chosen in order to represent a large variety of different atmospheric conditions, including trace-gas and aerosol free atmospheres as well as moderate to high trace gas and aerosol loads up to cloudy and foggy conditions. The shapes of the different profiles include near-surface box profiles, profiles exponentially decreasing with altitude, uplifted profiles with a Gaussian shape, as well as profiles without trace gases and aerosols, respectively. Additionally, the trace gas scenarios include two NO_2 profiles measured with a balloon sonde during the CINDI-2 campaign held in the Netherlands in