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Supplement of

Tropospheric NO_2 measurements using a three-wavelength optical parametric oscillator differential absorption lidar

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Supplementary Material

S.1 Two-wavelength DIAL retrieval equation

Table s.1. Extinction and backscatter of molecule and aerosol for wavelengths of λ_1 *and* λ_2.

wavelength	Molecular backscattering	Aerosol backscattering	Molecular extinction	Aerosol extinction
$\boldsymbol{\lambda_1}$	$\left(\frac{\lambda_1}{\lambda_2}\right)^{-4}\boldsymbol{\beta_m}$	$\left(\frac{\lambda_1}{\lambda_2}\right)^{-e}\boldsymbol{\beta_a}$	$\left(\frac{\lambda_1}{\lambda_2}\right)^{-4}\boldsymbol{\alpha_m}$	$\left(\frac{\lambda_1}{\lambda_2}\right)^{-e}\boldsymbol{\alpha_a}$
$\boldsymbol{\lambda_2}$	$\boldsymbol{\beta_m}$	$\boldsymbol{\beta_a}$	$\boldsymbol{\alpha_m}$	$\boldsymbol{\alpha_a}$

The two elastic lidar equations can be expressed as:

$$X(\lambda_1, Z) = C_1 \frac{\left[\left(\frac{\lambda_1}{\lambda_2}\right)^{-4}\beta_m(Z)+\left(\frac{\lambda_2}{\lambda_1}\right)^{-e}\beta_a(Z)\right]}{Z^2} \exp\left\{-2\int_0^Z \left[\left(\frac{\lambda_1}{\lambda_2}\right)^{-4}\alpha_m(z) + \left(\frac{\lambda_2}{\lambda_1}\right)^{-e}\alpha_a(z) + \sigma_N(\lambda_1, z)N_N(z) + O_{abs}(\lambda_1, z)\right] dz\right\} \quad \text{(s.1)}$$

$$X(\lambda_2, Z) = C_2 \frac{[\beta_m(Z)+\beta_a(Z)]}{Z^2} \exp\{-2\int_0^Z [\alpha_m(z) + \alpha_a(z) + \sigma_N(\lambda_2, z)N_N(z) + O_{abs}(\lambda_2, z)]dz\} \quad \text{(s.2)}$$

where X is the lidar signal; C_1 and C_2 are lidar constants; the subscripts a and m represent aerosol, and molecule, respectively; σ_N is the absorption cross section for the gas of interest; N_N is the molecular density of the gas of interest; O_{abs} is absorption of gases other than the gas of interest and z is the altitude. The molecular density of the gas of interest can be obtained after taking ratio of Eq. (1) to Eq. (2).

NO_2 density retrieval equation can be expressed as Eq. (3):

$$N_N(Z) = \frac{\frac{1}{2}\times\frac{d}{dz}\left[ln\frac{X(\lambda_1,Z)}{X(\lambda_2,Z)}\right]-AED(z)-MED(z)-OAD(z)-B(z)}{\Delta\sigma_N} \quad \text{(s.3)}$$

$$\Delta\sigma_N = \sigma_N(\lambda_2) - \sigma_N(\lambda_1) \quad \text{(s.4)}$$

$$B(z) = \frac{1}{2}\frac{d}{dz}\left[ln\frac{\left(\frac{\lambda_1}{\lambda_2}\right)^{-4}\beta_m(Z)+\left(\frac{\lambda_1}{\lambda_2}\right)^{-e}\beta_a(Z)}{\beta_m(Z)+\beta_a(Z)}\right] \quad \text{(s.5)}$$

$$AED(z) = \left[1 - \left(\frac{\lambda_1}{\lambda_2}\right)^{-e}\right]\alpha_a(Z) \quad \text{(s.6)}$$

$$MED(z) = \left[1 - \left(\frac{\lambda_1}{\lambda_2}\right)^{-4}\right]\alpha_m(Z) \quad \text{(s.7)}$$

$$OAD(z) = O_{abs}(\lambda_2, z) - O_{abs}(\lambda_1, z) \quad \text{(s.8)}$$

S.2 Units for all variables

Table s.2. Units for all variables

Items	Description	Unit
C1, C2,C3	lidar constant	constant (number)
α	extinction coefficient	km^{-1}
β	backscattering coefficient	$km^{-1}sr^{-1}$
λ	wavelength	nm
σ	absorption cross section	$cm^2molecule^{-1}$
Z, z	altitude	km
X	lidar range-corrected signal	mv
N	number density	$molecule/cm^3$
S	lidar ratio	sr^{-1}
U	Relative uncertainty	%