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Supplement of

Optical characterization of pure pollen types using a multi-wavelength Raman polarization lidar

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1 Particle linear depolarization ratio calculation (Eq.3 in the manuscript)

We follow the detailed calculations in Tesche et al. 2009.

Lidar-derived particle depolarization ratio (δ_{particle}) can be expressed as the ratio of cross- ($\beta^{\perp}$) and parallel- ($\beta^{/\!/}$) polarized particle backscatter coefficient:

$$\delta_{\text{particle}} = \frac{\beta^{\perp}_{\text{pollen}}+\beta^{\perp}_{\text{background}}}{\beta^{/\!/}_{\text{pollen}}+\beta^{/\!/}_{\text{background}}} \tag{S1}$$

The particle backscatter coefficient β_{particle} is the sum of cross- and parallel-polarized particle backscatter coefficient of both pollen and background aerosols:

$$\beta_{\text{particle}} = \beta^{\perp}_{\text{pollen}} + \beta^{\perp}_{\text{background}} + \beta^{/\!/}_{\text{pollen}} + \beta^{/\!/}_{\text{background}} \tag{S2}$$

The depolarization ratio of one particle type can be defined as:

$$\delta_x = \frac{\beta^{\perp}_x}{\beta^{/\!/}_x} \tag{S3}$$

The index *x=pollen* or *background* denotes the contribution of pollen or background particles, respectively. We can use the following relationships mathematically:

$$\beta_x = \beta^{\perp}_x + \beta^{/\!/}_x \tag{S4}$$

$$\beta^{/\!/}_x = \frac{\beta_x}{1+\delta_x} \tag{S5}$$

$$\beta^{\perp}_x = \frac{\beta_x \delta_x}{1+\delta_x} \tag{S6}$$

We replace equations S5 and S6 in equation S2, the particle linear depolarization ratio can be then calculated using the particle backscatter coefficients (β_{pollen} and $\beta_{\text{background}}$) and the depolarization ratios of both particle types (δ_{pollen} and $\delta_{\text{background}}$):

$$\delta_{\text{particle}} = \frac{\frac{\beta_{\text{pollen}} * \delta_{\text{pollen}}}{\delta_{\text{pollen}}+1}+\frac{\beta_{\text{background}} * \delta_{\text{background}}}{\delta_{\text{background}}+1}}{\frac{\beta_{\text{pollen}}}{\delta_{\text{pollen}}+1}+\frac{\beta_{\text{background}}}{\delta_{\text{background}}+1}} \tag{3}$$

2 Relationship of $\text{Å}_{\text{particle}}$ and χ_{pollen} (Eq.5 in the manuscript)

Two aerosol populations, pollen (depolarizing) and background (non-depolarizing) aerosols are considered. The backscatter coefficient of the total particles is the sum of backscatter coefficient of both pollen and background aerosols:

$$\beta_{\text{particle}}(\lambda_1) = \beta_{\text{pollen}}(\lambda_1) + \beta_{\text{background}}(\lambda_1) \tag{S7a}$$

$$\beta_{\text{particle}}(\lambda_2) = \beta_{\text{pollen}}(\lambda_2) + \beta_{\text{background}}(\lambda_2) \tag{S7b}$$

Similar as Eq.2 in the manuscript, the backscatter-related Ångström exponent (Å) can also be expressed in this equation:

$$\left(\frac{\lambda_1}{\lambda_2}\right)^{-\text{Å}_x(\lambda_1,\lambda_2)} = \frac{\beta_x(\lambda_1)}{\beta_x(\lambda_2)} \tag{S8}$$

The index *x=pollen, background* or *particle* denotes the backscatter-related Ångström exponent of pollen, background or total particles.

We replace the top part of right side of Eq.S8 with *x= particle* with Eq.S7. And further use expression of $\beta_{\text{pollen}}(\lambda_2)$ and $\beta_{\text{background}}(\lambda_2)$ to replace the $\beta_{\text{pollen}}(\lambda_1)$ and $\beta_{\text{background}}(\lambda_1)$ in Eq.S7a, based on Eq.S8. Thus we have:

$$\left(\frac{\lambda_1}{\lambda_2}\right)^{-\text{Å}_{\text{particle}}(\lambda_1,\lambda_2)} = \frac{\left(\frac{\lambda_1}{\lambda_2}\right)^{-\text{Å}_{\text{pollen}}(\lambda_1,\lambda_2)} * \beta_{\text{pollen}}(\lambda_2)+\left(\frac{\lambda_1}{\lambda_2}\right)^{-\text{Å}_{\text{background}}(\lambda_1,\lambda_2)} * \beta_{\text{background}}(\lambda_2)}{\beta_{\text{particle}}(\lambda_2)} \tag{S9}$$

After replacing $\beta_{\text{background}}(\lambda_2)$ with Eq.S7b, the equation can be expressed as:

$$\left(\frac{\lambda_1}{\lambda_2}\right)^{-\text{Å}_{\text{particle}}(\lambda_1,\lambda_2)} = \frac{\left(\left(\frac{\lambda_1}{\lambda_2}\right)^{-\text{Å}_{\text{pollen}}(\lambda_1,\lambda_2)} - \left(\frac{\lambda_1}{\lambda_2}\right)^{-\text{Å}_{\text{background}}(\lambda_1,\lambda_2)}\right) * \beta_{\text{pollen}}(\lambda_2) + \left(\frac{\lambda_1}{\lambda_2}\right)^{-\text{Å}_{\text{background}}(\lambda_1,\lambda_2)} * \beta_{\text{particle}}(\lambda_2)}{\beta_{\text{particle}}(\lambda_2)} \quad \text{(S10)}$$

Using the definition of pollen backscatter contribution (Eq.4 in the manuscript), a linear relationship between $\left(\frac{\lambda_1}{\lambda_2}\right)^{-\text{Å}_{\text{particle}}(\lambda_1,\lambda_2)}$ and $\chi_{\text{pollen}}(\lambda_2)$ can be retrieved for the wavelength pair (λ_1, λ_2):

$$\left(\frac{\lambda_1}{\lambda_2}\right)^{-\text{Å}_{\text{particle}}(\lambda_1,\lambda_2)} = \left(\left(\frac{\lambda_1}{\lambda_2}\right)^{-\text{Å}_{\text{pollen}}(\lambda_1,\lambda_2)} - \left(\frac{\lambda_1}{\lambda_2}\right)^{-\text{Å}_{\text{background}}(\lambda_1,\lambda_2)}\right)\chi_{\text{pollen}}(\lambda_2) + \left(\frac{\lambda_1}{\lambda_2}\right)^{-\text{Å}_{\text{background}}(\lambda_1,\lambda_2)} \quad \text{(S11a)}$$

A similar formulate is found for the wavelength pair (λ_2, λ_1) when considering $\chi_{\text{pollen}}(\lambda_1)$:

$$\left(\frac{\lambda_2}{\lambda_1}\right)^{-\text{Å}_{\text{particle}}(\lambda_1,\lambda_2)} = \left(\left(\frac{\lambda_2}{\lambda_1}\right)^{-\text{Å}_{\text{pollen}}(\lambda_1,\lambda_2)} - \left(\frac{\lambda_2}{\lambda_1}\right)^{-\text{Å}_{\text{background}}(\lambda_1,\lambda_2)}\right)\chi_{\text{pollen}}(\lambda_1) + \left(\frac{\lambda_2}{\lambda_1}\right)^{-\text{Å}_{\text{background}}(\lambda_1,\lambda_2)} \quad \text{(S11b)}$$

3 Supplementary figures

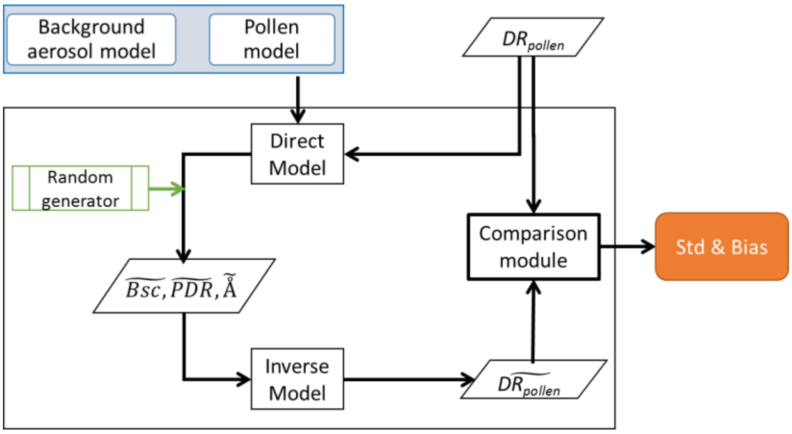

Figure S 1. Block diagram of the end-to-end simulator. Detail description is in section 3.

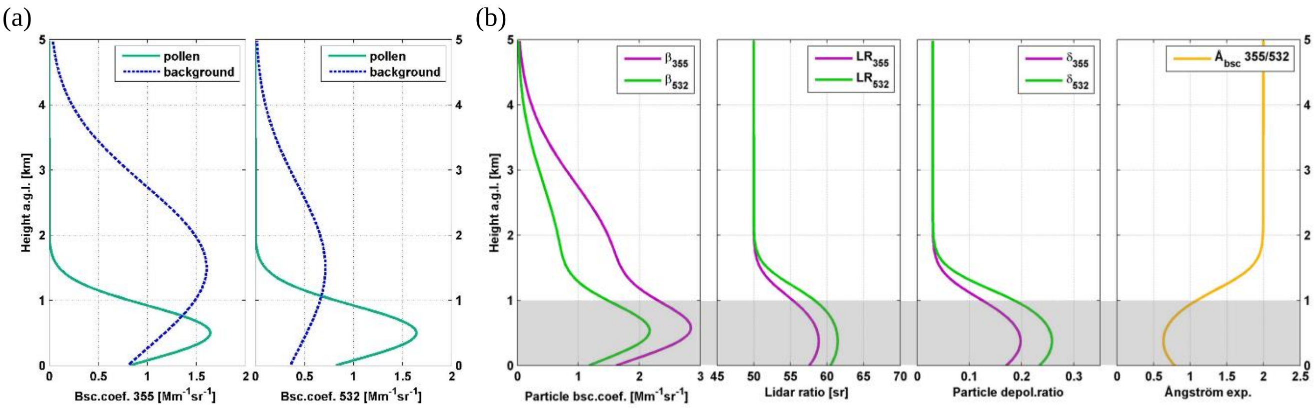

Figure S 2. Simulated profiles from the direct model, using parameters given in Table 1. The values of optical depth of both background and pollen aerosol layers are selected as 0.1 for this case. (a) Simulated backscatter coefficient of pollen (in dark green lines) and background (in dotted blue lines) aerosol layers at 355 nm or 532 nm. (b) Simulated particle backscatter coefficient, lidar ratio, particle depolarization ratio of the total aerosols at 355 nm (in purple) or 532 nm (in green), and backscatter-related Ångström exponent at 355-532 nm. The grey area indicates the defined pollen layer.

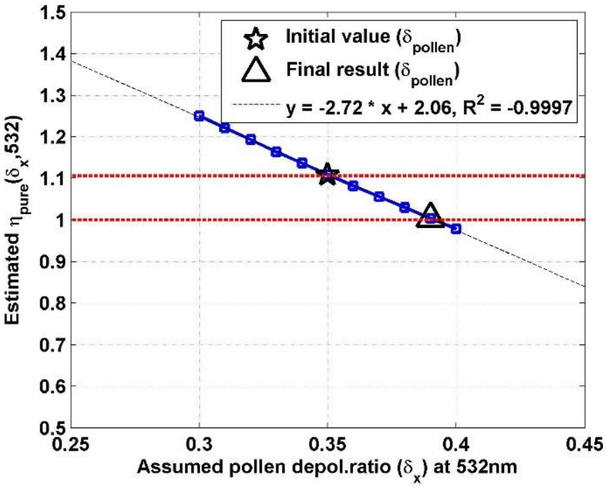

Figure S 3. Estimated parameter η_{pure} against the related assumed pollen depolarization ratio δ_x at 532 nm. η_{pure} is the $\eta(\chi_{pollen})$ value for the pure pollen (100 % pollen in the observed aerosol particle population, $\chi_{pollen} = 1$), where η is a parameter (Eq.6) using backscatter-related Ångström exponent between 355 and 532 nm (Å). Linear regression line is drawn by black dotted line, with fitting equation shown. The correlation coefficient (R^2) value is also given. The initial values (shown by the black pentagram) are 0.35 for pollen depolarization ratio and 0.25 for $Å_{pollen}$ (i.e. 1.11 for η_{pure}). As we assume $\hat{\eta}_{pure}$=1 (i.e. $\widehat{Å}_{pollen}$=0) in the inverse model for the retrieval, the final result of 0.39 for pure pollen is found (by the black triangle).

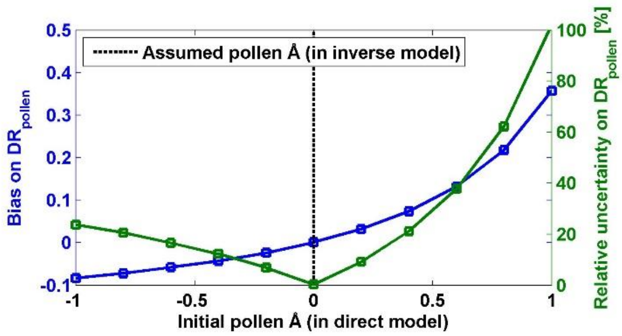

Figure S 4. Examples of estimated bias and relative uncertainty on retrieved pollen depolarization ratio (DR_{pollen}) against the initial values of backscatter-related Ångström exponent between 355 and 532 nm (Å) for pollen. The pollen Å is assumed as 0 (i.e. $\widehat{Å}_{pollen}$=0) in the inverse model for the calculation (shown by black dotted line).

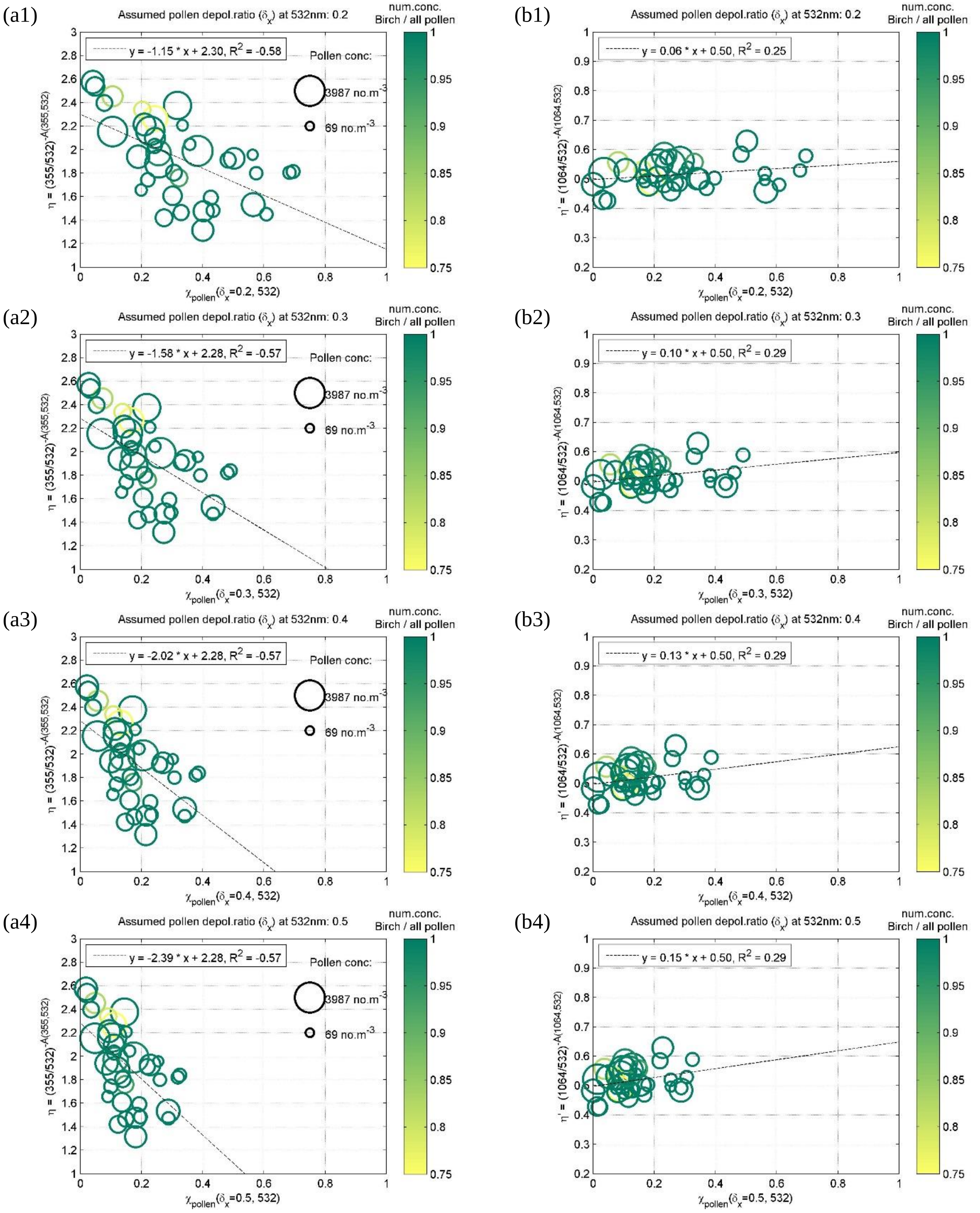

Figure S 5. Mean values of the parameter $\eta(\lambda_1, \lambda_2) = \left(\frac{\lambda_1}{\lambda_2}\right)^{-\text{Å}_{\text{particle}}(\lambda_1,\lambda_2)}$ against pollen backscatter contribution $\chi_{\text{pollen}}(\lambda_2)$ at the wavelength of λ_2 inside the pollen layers during IPP-1. The wavelength pairs (λ_1, λ_2) are selected as (355,532) in (a1-a4), and (1064,532) in (b1-b4). The two parameters η (a), and η′ (b) are functions (Eq.6) of the backscatter-related Ångström exponent between 355 and 532 nm or between 532 and 1064 nm, for the total particle backscatter coefficients. The pollen depolarization ratio at 532 nm (δ_x) is assumed to be 0.2, 0.3, 0.4 or 0.5 (from top to bottom). Linear regression lines are drawn by dotted lines, with fitting equations shown (Eq.5 or 8). The correlation coefficient (R^2) is also given. The size denotes the total pollen concentrations measured by the Burkard sampler on roof level; the colour represents the number concentration of the dominant pollen (birch) against the total pollen number concentration.

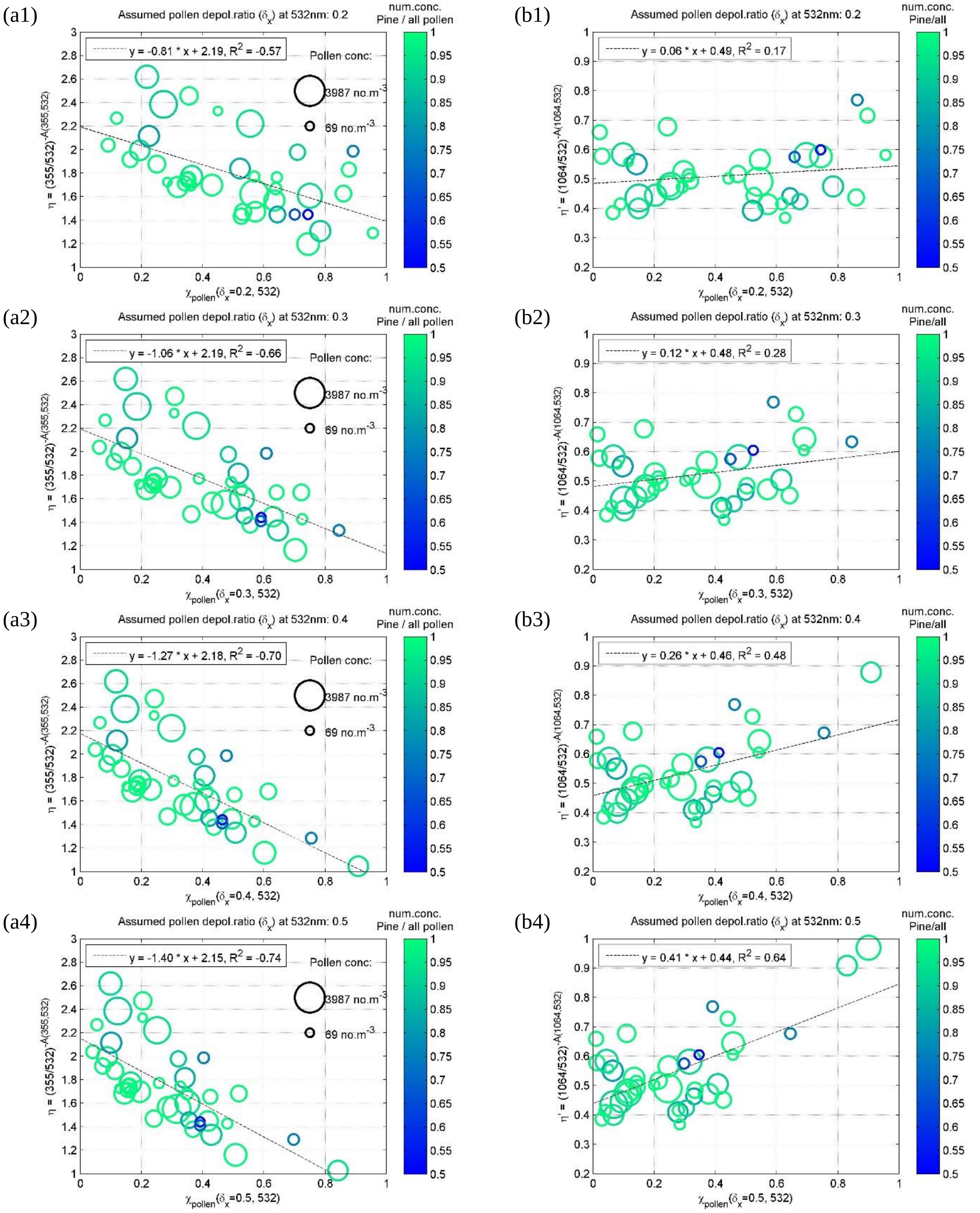

Figure S 6. Similar as Fig. S5, but for IPP-3 (pine is dominant pollen).

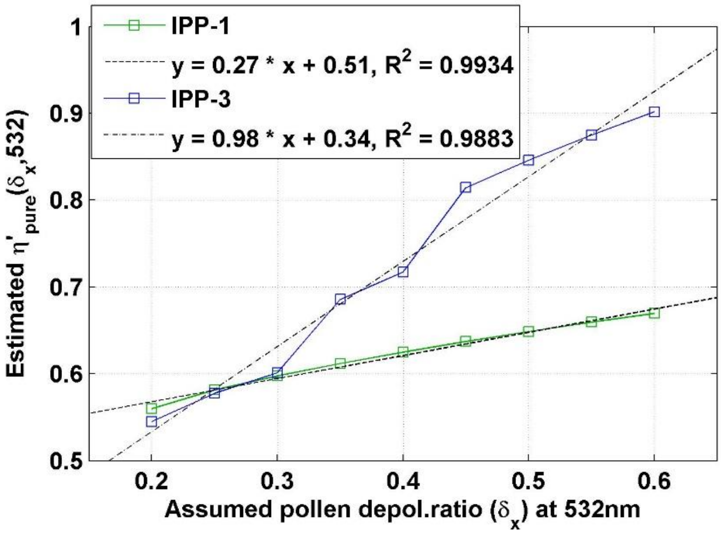

Figure S 7. Similar as Fig. 12, but for the estimated $\eta'_{\mathbf{pure}}(\boldsymbol{\delta_x}, \mathbf{532})$ against the related assumed pollen depolarization ratio $\boldsymbol{\delta_x}$ at 532 nm for IPP-1 (in green) and IPP-3 (in blue). η' is a function of backscatter-related Ångström exponent between 532 and 1064 nm (Eq.6), and $\eta'_{\mathbf{pure}}$ is the estimated η' value for $\boldsymbol{\chi}_{\mathbf{pollen}}(\boldsymbol{\delta_x})$=1 (i.e. pollen contribution in the observed aerosol particle population is 100%) (Eq.8).

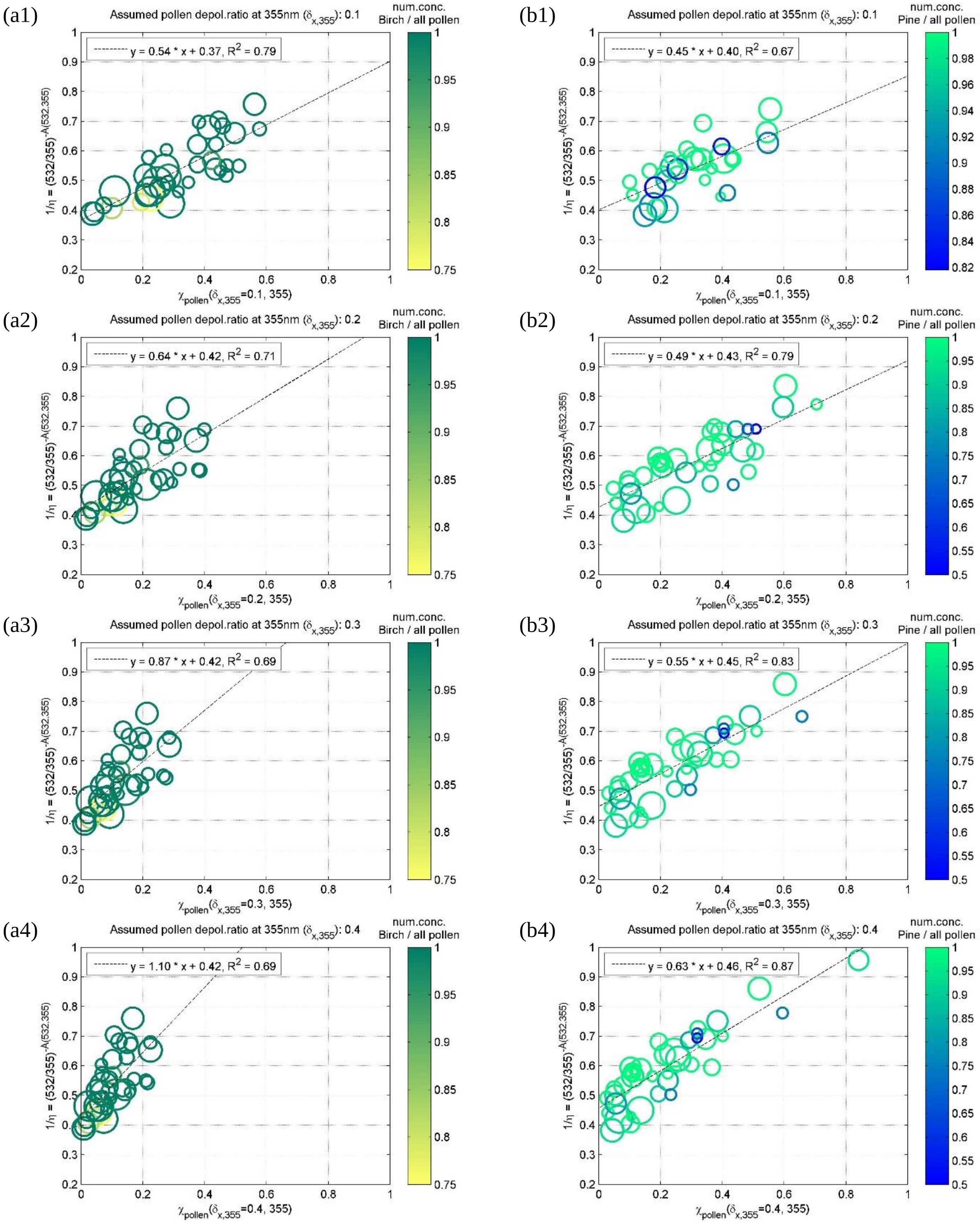

Figure S 8. Mean values of the parameter $\frac{1}{\eta}$ against pollen backscatter contribution at 355 nm $\chi_{\mathbf{pollen}}(355)$ inside the pollen layers during IPP-1 (a1-a4), or IPP-3 (b1-b4). The parameter $\frac{1}{\eta}$ is a function ($\frac{1}{\eta} = \left(\frac{532}{355}\right)^{-\text{Å}_{\text{particle}}\,(532,355)}$ in Eq.6) of the backscatter-related Ångström exponent between 355 and 532 nm for the total particle backscatter coefficients. The pollen depolarization ratio at 355 nm ($\delta_{x,355}$) is assumed to be 0.1, 0.2, 0.3, or 0.4 (from top to bottom). Linear regression lines are drawn by dotted lines, with fitting equations shown (Eq.5 or 8). The correlation coefficient (R^2) is also given. The size denotes the total pollen concentrations measured by the Burkard sampler on roof level; the colour represents the number concentration of the dominant pollen (birch for (a) and pine for (b)) against the total pollen number concentration.