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Spaceborne imaging spectrometry of methane plumes: Quantifying the benefit of aerosol lidar

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Key Points:

- Aerosol extinction leads to biased methane enhancements (difference between plume and background XCH₄) if not accounted for by the retrieval
- The aerosol profile from lidar may constrain the retrieval, but it would miss the aerosol profile for the plume or the background
- Using end-to-end simulation in Monte Carlo mode suggests: A single profile from the swath center may still reduce the enhancement bias

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19 Abstract (250 words max)

20 Column averaged mixing ratios of trace gases, such as methane (XCH_4), from spaceborne
21 pushbroom spectrometers can be used to detect corresponding plumes and retrieve
22 enhancements (ΔXCH_4), i.e., the difference between plume and background XCH_4 . Over the
23 global dust belt, however, significant scattering by dust aerosols may cause biased XCH_4 that
24 may propagate into biased ΔXCH_4 . To correct this, a co-aligned lidar could measure the aerosol
25 profile in the middle of the scene (~20 km across track by 6 km along track). However, the
26 vertical aerosol distribution may vary between the plume, covering hundreds of meters, and
27 the background. Due to signal-to-noise requirements, only a single lidar profile per scene could
28 be acquired, so either the plume or the background aerosol layer would be missed. Aerosol-
29 induced bias would not be corrected equally for plume and background. A single lidar sounding
30 in the center of the swath may, therefore, not improve the ΔXCH_4 bias. To test this hypothesis,
31 end-to-end simulations in Monte Carlo mode were performed, simulating 2000 retrievals for
32 randomized aerosol profiles and observation conditions. The use of a scattering radiative
33 transfer model (RTM) constrained with aerosol profile data from a lidar at the swath center led
34 to a reduced mean absolute bias of ΔXCH_4 (~0.5 ppb vs. ~2.6 ppb for an unconstrained RTM),
35 with highest improvements for low albedo and strong plume/background albedo contrasts. This
36 suggests that a pushbroom-lidar doublet could enable more accurate mass fluxes from point
37 gas emitters, such as man-made greenhouse gas sources or volcanoes.

38

39 Plain Language Summary

40 To obtain methane enhancements (ΔXCH_4 , plumes) and mass emission rates from imaging
41 spectrometer retrievals, methane concentrations (XCH_4) at the plume and the background
42 pixels are measured. Their difference yields ΔXCH_4 . Soundings will be performed over
43 atmospheres containing substantial amounts of dust aerosol, which modify light propagation.
44 Both plume and background XCH_4 may thus be erroneous. This may cause erroneous ΔXCH_4 and
45 mass flux estimates. To account for aerosol extinction, for the first time, a co-aligned micro-
46 pulsed lidar, measuring the aerosol profile is proposed. But it could only deliver a single lidar
47 profile for the entire scene (20 km across track by 6 km along track). The lidar would either miss
48 the aerosol over the plume or the background. Therefore, XCH_4 could not be corrected equally
49 for both and a ΔXCH_4 error is expected to remain, making the benefit of a lidar questionable.
50 Assuming, a single aerosol type across the scene, smoothly transitioning between plume and
51 background, we show by simulating many measurements under different observation
52 conditions and aerosol distributions, that a single aerosol profile from lidar leads to fewer and
53 lower ΔXCH_4 errors, especially for low albedo, high aerosol density and if albedo strongly varies
54 between plume and background pixels.

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58 1 Introduction

59 Over the last two decades, a new generation of satellites-borne imaging spectrometers with
60 sub-kilometer pixel resolution emerged. This includes EnMAP (Guanter et al., 2015), EMIT
61 (Thorpe et al., 2023), MethaneSAT (Dandenault et al., 2020) or CarbonMapper (Duren et al.,
62 2025). These platforms can image and quantify atmospheric trace gas enhancements (plumes)
63 over small areas, in particular, those from point emitters, such as CO₂ or CH₄ from oil and gas
64 facilities. This enables direct quantification of fluxes of greenhouse gases (GHG) at a more
65 regional or even local scale, contributing to improved emission inventories and emission
66 factors.

67 Among these platforms are the first commercial EO missions such as GHGSat (Chan
68 Miller et al., 2024), and AIRMO-Space. The latter has the aim to launch a satellite constellation
69 of 12 MicroSat class spacecraft to detect and quantify methane plumes using pushbroom
70 hyperspectral imaging in the short wave infrared (SWIR) region in target mode, providing the
71 following data products: Level-2 (L2, images of CH₄ column averaged mixing ratios, XCH₄), Level-
72 3 (L3, plume masks with CH₄ enhancements, ΔXCH₄) and level-4 (L4, mass emission rates or
73 fluxes).

74 Following end-to-end simulations, the performance budget to reach the envisaged L4
75 sensitivity of 100 kgCH₄h⁻¹ from point emitters entails a target precision and bias of 16 ppbCH₄
76 and -0.5 to 3.3 ppbCH₄, respectively, comparable to missions with similar objectives (Noël et al.,
77 2024; MacLean et al., 2025).

78 The in-orbit-demonstration (IOD) will concentrate on oil and gas operations in areas in
79 North Africa, Middle East and Asia. Large extends of this region are located in the Dust Belt
80 (Milford et al., 2019; Tindan et al., 2023), where an aerosol optical depth (AOD) >0.1 is expected
81 at many locations all year around, based on AERONET data (Holben et al., 1998). Over these
82 areas, light scattering and absorption by aerosols may significantly modify the path of
83 downwelling and upwelling solar photons, leading to over- or underestimated trace gas column
84 densities, i.e., biased XCH₄ (Butz et al. 2012; Somkuti et al. 2025).

85 Proxy gas retrieval using XCO₂ (Frankenberg et al.; 2005, 2025; Krings et al., 2011;
86 Schepers et al., 2012; Staebell et al., 2021) is viable and considered for the current mission.
87 However, for areas where homogeneous XCO₂ cannot be assumed, including urban areas
88 (Klausner et al., 2020; Zhu et al., 2022), or near large geological CO₂ emitters (Di Martino et al.,
89 2024), we deem a physics-based retrieval, i.e. modeling aerosol extinction during the L2
90 retrieval, the most adequate. This requires a scattering forward radiative transfer model
91 (scattering RTM, SRTM), which in turn needs an aerosol parametrization, such as in the form of
92 aerosol layer height (ALH), aerosol layer width (ALW) and AOD (Dubovik et al., 2011; Butz et al.,
93 2012). ALH and ALW are commonly retrieved indirectly by measuring and inverting the Oxygen
94 A-band absorption, which, however, may entail uncertainties of the order of ~1 km (Jänicke et
95 al., 2023; Kim et al., 2024).

96 To achieve more direct and potentially more accurate measurements of the aerosol
97 profile, a dedicated micro aerosol lidar, co-aligned with the imaging spectrometer, is planned to
98 be part of the payload. Simulations for AOD >0.1 and a consequent lidar signal-to-noise ratio
99 (SNR) ≥ 3 yielded range uncertainties <200 m. The pulsed lidar will probe the entire

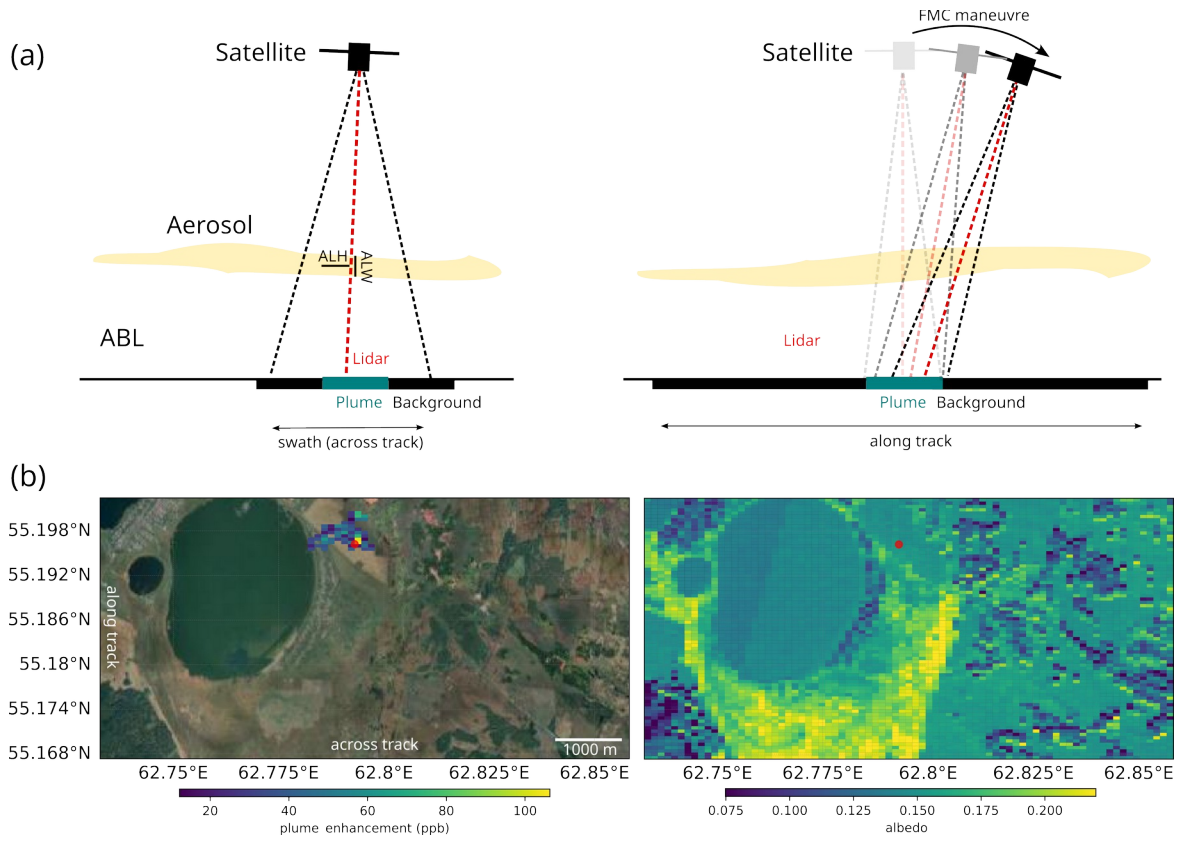
100 atmospheric column beneath the spectrometer, directly measuring the aerosol attenuated
101 backscatter profile, and thus the vertical aerosol distribution (ALH, ALW and potentially AOD).

102 The latter would constrain the SRTM, which would effectively improve vertical
103 sensitivity of the retrieval as quantified by the methane averaging kernel (AK), a diagnostic
104 metric calculated as $AK = GK$, where G is the gain vector (for CH_4 , in ppb per
105 photons/s/m²/sr/nm) and K the Jacobian matrix. The latter specifies the sensitivity of the
106 forward modeled radiance to a change in a state vector element at a given vertical atmospheric
107 layer of the RTM and thus the vertical retrieval sensitivity of the total column XCH_4 . It describes
108 how a perturbation of methane concentration in a given layer (relative to the prior) propagates
109 into the retrieved total column XCH_4 . Aerosol scattering would be allocated at the correct
110 altitude, improving the Jacobian and thus the AK. A SRTM constrained with accurate ALH and
111 ALW would therefore reduce misinterpretation in the L2 retrieval, particularly when aerosols
112 are not vertically co-located with trace gas accumulations, as it would help to differentiate
113 between a radiance perturbation due to aerosol or gas.

114 However, following lidar sensitivity analysis, to reach the minimum SNR, the lidar would
115 be integrating pulses during tens of seconds, while a forward motion compensation (FMC)
116 maneuver is executed from a zenith angle of about -15° to 15° (Fig. 1a). The corresponding
117 scene length (along track) would be ~ 6 km and the scene width (across track) would be ~ 20 km.

118 Full 2D end-to-end (E2E) simulations using $100 \text{ kgCH}_4\text{h}^{-1}$ plumes from large eddy
119 simulations (LES) suggest numerous scenarios where either the background or the plume would
120 be outside the lidar field of view (FOV) of ~ 130 m (e.g., Fig. 1b). The lidar would basically
121 perform a slice-scanning in the swath center and along the flight track, yielding an average
122 measure of the aerosol profile along track. It would miss aerosol left and right of its FOV, i.e.,
123 over the background or plume pixels, whichever are not beneath the lidar. This leads to the
124 main question of the paper: The potential benefit of aerosol profile from such a lidar sounding.

125 The latter is critically questioned in the following, leading to the hypothesis. In the
126 method section, the simulation scheme used to test the hypothesis is detailed. In the result
127 section, the simulation results are presented and discussed.



129

130 **Figure 1.** Observation situation. (a) Schematic view of measurement geometry with a staring
 131 lidar co-aligned with the push broom spectrometer. In this example the background XCH_4 is off
 132 the center swath, whilst the XCH_4 enhancement (plume) is at the center of the swath. Left:
 133 across track view. ALH and ALW are measured in the swath center. Right: Along track view
 134 depicting the forward motion compensation (FMC). ALH and ALW represent average values
 135 over the probed track. ABL depicts the atmospheric boundary layer. Figure not to scale. (b)
 136 Simulated scene (zoom) with retrieved L3 enhancements from a LES plume for $100 \text{ kgCH}_4\text{h}^{-1}$
 137 flux and 2 ms^{-1} wind speed (simulation with enhancement form a LES plume) over satellite
 138 imagery (Source of background tiles: Google). Easting is aligned with across-track. The red dot
 139 depicts the randomly chosen source location. Right: Albedo from Sentinel-2 of the same
 140 location. Re-gridded to GSD of instrument (50 m along track and 100 m across track).

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143 3 Material and Methods

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145 3.1 Null hypothesis

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147 The key quantity of interest for this mission is ΔXCH_4 , i.e., the difference between plume and
 148 background XCH_4 (L3, enhancement). In an idealized model, assuming a plume and a

149 background pixel with equal surface reflectivity, atmosphere and solar irradiance, respectively,
 150 the retrieved, aerosol-biased XCH_4 for plume and background could be written as

$$151 \quad XCH_{4,pl,based} = XCH_{4,pl} + G(y_{aero,pl} - y_{aero,lidar}),$$

153
 154 and

$$155 \quad XCH_{4,bg,based} = XCH_{4,bg} + G(y_{aero,bg} - y_{aero,lidar}). \quad (1)$$

157 Let G be the (spectrally dependent) gain vector and $y_{aero,pl}$ and $y_{aero,bg}$, the fraction of the
 158 radiance caused by aerosol scattering over plume and background, both unexplainable by the
 159 RTM, and $y_{aero,lidar}$ the radiance explained by a SRTM constrained with an aerosol profile from
 160 the swath center. $X_{CH_4,pl}$ and $X_{CH_4,bg}$ are the mixing ratios if no aerosol was present. In the
 161 retrieval used here, the enhancement (L3) would then be

$$162 \quad \Delta XCH_4 = X_{CH_4,pl,true} - X_{CH_4,bg,true} + \varepsilon_{\Delta XCH_4}, \quad (2)$$

163
 164 with the L3 error, i.e., the aerosol-induced enhancement bias,

$$165 \quad \varepsilon_{\Delta XCH_4} = G(y_{aero,pl} - y_{aero,bg}). \quad (3)$$

166
 167 $y_{aero,lidar}$ constrained by a single aerosol profile canceled in Eq. (2) and any existent bias, Eq. (3),
 168 remains, i.e., is not corrected. From Eq. (3) it also follows that if the aerosol induced bias of
 169 plume and background are equal, such as when the aerosol profile is the same for plume and
 170 background, the bias would cancel. The effect of a lidar would be nullified in both cases.

171 However, the above model oversimplifies reality, for instance, in assuming equal
 172 observation conditions, spectral gain for plume and background and linearity of the retrieval
 173 response. A single aerosol profile for the scene, such as retrieved from lidar, could still give a
 174 smaller enhancement bias compared to not correcting for aerosol scattering. To assess this, the
 175 following null hypothesis was tested: A single measured aerosol profile per scene does not
 176 reduce the ΔXCH_4 bias $\varepsilon_{\Delta XCH_4}$.

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180 3.2 Simulation approach

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 182 To test the null hypothesis, end-to-end simulations were performed using a Monte Carlo
 183 simulation (MCS) approach. A single aerosol layer was assumed, smoothly varying in height
 184 (ALH), width (ALW) and aerosol number density (parametrized as AOD), over the scene,
 185 including the enhancement (plume). MCS using full scenes with hundreds of across-track and
 186 along track pixels, respectively, would be computationally expensive. Moreover, it would add
 187 another layer of complexity in identifying statistical difference in L3 bias with/without SRTM.
 188 Therefore, the smallest possible scene was simulated: a “plume pixel”, containing the enhanced
 189 methane column, and a “background pixel”, which contains only the background methane
 190
 191

192 column. Although a simplification, this two-pixel scene is deemed to conserve the essential
 193 radiative transfer physics that governs the biases in L2 and L3. The plume was assumed to be at
 194 the swath center, probed by the lidar, the background pixel was off the swath center and not
 195 probed by the lidar (as in Fig. 1b). This is arbitrary and reversing this assumption would not
 196 reduce generality.

197 Considering a L4 sensitivity requirement of $\sim 100 \text{ kgCH}_4\text{h}^{-1}$, the true enhancement
 198 $\Delta XCH_{4,true}$ was estimated using a simple slab mass balance model (Gerilowski et al., 2011),
 199 linking methane enhancement and plume transport speed to mass emission rate (flux). The
 200 column enhancement was assumed to be equally spread over a plume width of 160 m.
 201 Assuming a wind speed of 1 ms^{-1} entails a column enhancement of $\sim 6.5 \times 10^{21} \text{ molec.m}^{-2}$ or
 202 $\sim 1.6\%$, assuming a background XCH_4 of 1900 ppb. This corresponds to a ~ 30 ppb enhancement
 203 adding to an assumed 1900 ppb background methane mixing ratio.

204 A complete MCS run consisted of the following steps:

- 205 1. Simulate top-of-atmosphere (TOA) radiance for the plume pixel with plume aerosol
 206 profile, XCH_4 1930 ppb
- 207 2. Simulate TOA radiance for the background pixel with background aerosol profile, XCH_4
 208 1900 ppb
- 209 3. Retrieve XCH_4 (L2) for plume pixel using the SRTM
- 210 4. Retrieve XCH_4 for background pixel using the SRTM
- 211 5. Retrieve XCH_4 for plume pixel using the NSRTM
- 212 6. Retrieve XCH_4 for background pixel using the NSRTM
- 213 7. Compute $\Delta XCH_{4,SRTM}$ and $\Delta XCH_{4,NSRTM}$
- 214 8. For both, compute the L3 bias as

$$215 \quad \varepsilon_{\Delta XCH_4} = \Delta XCH_{4,SRTM} - \Delta XCH_{4,true} = (XCH_{4,pl} - XCH_{4,pl,true}) - (XCH_{4,bg} - XCH_{4,bg,true}) \quad (4)$$

216

217 Equation (4) shows that the L3 bias is the difference between the L2 bias of plume and
 218 background.

219 The steps outline above were executed for 2000 random measurements (random
 220 draws), of observation conditions (OC, albedo and solar and viewing azimuth and zenith angles)
 221 and aerosol profiles, parameterized by ALH, ALW, AOD. Henceforth, a “measurement” refers to
 222 one of the 2000 retrievals.

223 As an accuracy metric, which assesses the statistical significance of using a constrained
 224 SRTM over a NSRTM, the mean absolute error (MAE) of the enhancement was computed as
 225

$$226 \quad MAE = \frac{1}{2000} \sum_{i=1}^{2000} |\varepsilon_{\Delta XCH_4}|_i \quad (5)$$

227

228

229 3.3 End-to-End simulator overview

230

231 In the following, the end-to-end simulator (E2E) used to simulate TOA radiance and retrieve
 232 XCH_4 for the push broom spectrometer is briefly described (Fig. 2a). Table 1 summarizes the key

233 parameters used. Text 1 in the supporting information gives more details on the NSRTM, SRTM,
234 L2 retrieval and aerosol parametrization.

235 Given a set of OC and aerosol profile (AOD, ALH, ALW), for a given pixel, the scene
236 generation module (SGM) calculates a TOA spectra using the SRTM, accounting for absorption
237 of H₂O, CO₂ and CH₄ (Table 1) and assuming transported mineral dust aerosol across the whole
238 scene (Sugimoto et al., 2020; Ratcliffe et al., 2024). This aerosol type was deemed
239 representative for the mission's region of interest. True ALH, ALW and AOD (Table 1) are
240 representative values adopted from the AERONET observational network (Holben et al., 1998).
241 The aerosol type specific single scattering albedo and scattering phase function (Fig. S3 in the
242 supporting information) were taken from the OPAC catalogue (Hess et al., 1998).

243
244 **Table 1.** Push broom and E2E simulation parameters and aerosol scenario used for all
245 simulations with default AOD, ALH and ALW.

Parameter type	Value
Spectral range for SGM and L2	1590 nm - 1670 nm
Spectral resolution (ISRF)	0.9 nm (FWHM)
Ground pixel size	50 m along tracking 100 m across track
Swath width	20 km
Sensor altitude	500 km
True XCH ₄ background	1900 ppb
True XCH ₄ plume	1930 ppb
True XCO ₂ (plume and background)	425 ppm
P, T, spec. humidity	US-Standard Atmosphere
Aerosol type	Transported mineral dust
AOD	0.21
ALH	2500 m
ALW	750 m
Single scattering albedo @ 1550 nm	0.925
Mie scattering 4 π phase function at 150° scattering angle	0.248

246
247 Since the SNR of the lidar is relatively low, AOD was assumed not retrievable from the
248 lidar signal, but instead was assumed to be known reasonably well from observational
249 networks, such as AERONET. This was mimicked by setting the retrieval AOD used in the L2
250 forward modeling to the mean AOD between plume and background pixel for all MCS runs. A
251 corresponding sensitivity test assessing the effect of the AOD knowledge error is provided in
252 the supporting information, text S2.

253 To simulate a realistic, smoothly varying true aerosol layer between plume and
254 background (Fig. 1), the parameters of the background aerosol layer (i.e., ALW, ALH, AOD) were
255 drawn from a random distribution (mean/STD: 2500 m/700 m, 750 m/400 m, 0.21/0.13).

256 Smoothly correlated ALW, ALH and AOD for the plume pixel were simulated by perturbing the
257 background aerosol parameters by a random number with mean 1 and STD of 0.1 (10%). Figure
258 S2 (supporting information) shows example distributions for all parameters. Figure 2b shows an
259 example aerosol profile as used in the SGM.

260 To avoid correlation, mimicking random differences in plume/background albedo (e.g.,
261 forest/crop/rock/urban boundaries, Fig. 1b), the plume and background albedo values were
262 drawn from two different random distributions. Viewing zenith angles (VZA) were different for
263 plume and background, but drawn from the same random distribution, i.e., not independent.
264 While random, solar zenith/azimuth (SZA, SAA) and viewing azimuth angles (VAA) were the
265 same for plume and background. ALH and ALW were clipped to a minimum of 300 and 200 m,
266 respectively. AOD was clipped to an interval of [0.02, 1], VZA, VAA, SZA, SAA were clipped to
267 [0°,20°], [0°,360°], [0°,70°], [0°, 360°], respectively. Albedo values were clipped to [0.1, 0.8].

268 The L0 radiance for each pixel is input to an instrument model (Fig. 2a), which simulates
269 the measurement process. In the configuration used here, to isolate the effect of aerosol
270 scattering, no instrument noise and no instrument errors were simulated. Only the degradation
271 in spectral resolution was simulated by convolving the TOA radiance with a Gaussian
272 instrumental spectral response function (ISRF), leading to simulated raw spectra (Level-0, L0)
273 for each pixel. These are passed to the L0-L1b module, which converts them to instrument
274 corrected and calibrated radiance spectra (L1b, Fig. 2a).

275 In the L2 processor, the L1b spectrum is fitted to a forward modeled radiance spectrum,
276 using either the SRTM or the NSRTM, resulting in column averaged mixing ratios XCH_4 (Fig. 2a).
277 Both the L1b-L2 and the SGM module use the same SRTM.

278

279 3.4 Lidar simulation

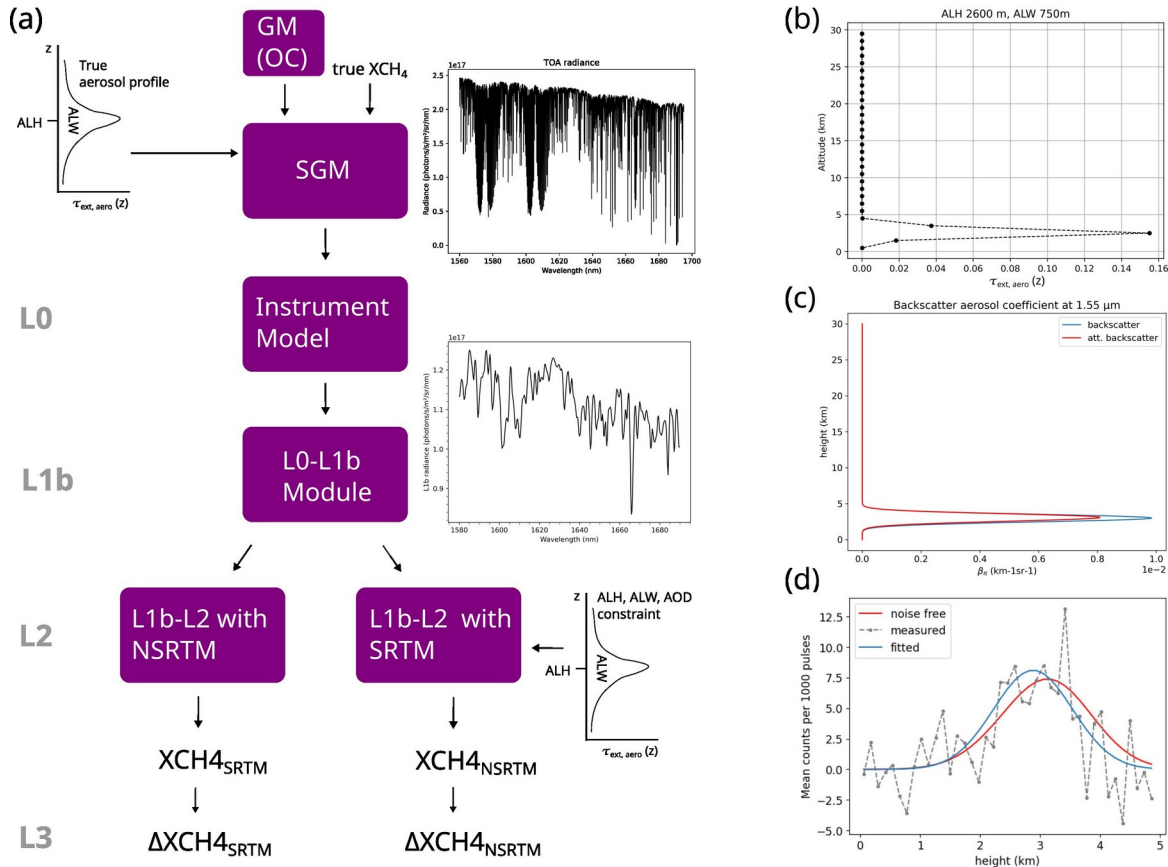
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281 The sole aim of the lidar simulation was to obtain a realistic noise (uncertainty) estimate for
282 lidar-retrieved ALH and ALW. Design parameters, such as repetition rate, optical power,
283 aperture, receiver bandwidth as well as the lidar simulator itself are out of scope and will be
284 detailed in a separate article. We assume here that the lidar can yield $SNR \geq 3$. The lidar
285 simulation evaluates the attenuated backscatter (Fig. 2c), which results from the same aerosol
286 profile as for SGM (e.g., Fig. 2b, but fixed AOD 0.21, ALH 3 km and ALW 750 m) and the lidar
287 ratio. This is followed by a detector module.

288 Noise was calculated as the contributions of signal, solar background and dark counts of
289 shot noise. The TOA radiance from the E2E SGM module was used to simulate solar background
290 photon noise as part of the SNR calculation for the lidar signal. The SNR was determined by the
291 desired range resolution and the measurement time for signal and background. The noise was
292 added to the expected signal based on the SNR profile. The resulting SNR was 4.

293 ALH and ALW were retrieved by fitting a Gaussian curve to the noisy lidar signal, Fig. 2d.
294 To assess uncertainty (mean and standard deviation, STD) of ALH and ALW, this procedure was
295 repeated in Monte Carlo manner with 1000 realizations of random noise, all associated with
296 SNR 4. This yielded ALH 3.09 ± 0.126 km and ALW 0.736 ± 0.144 km (uncertainties are sample
297 STD). The corresponding relative errors were adopted as representative lidar measurement
298 uncertainties of ALH and ALW during L2 retrieval by multiplying them with the true ALH and
299 ALW for the plume (swath center).

300 For simplicity, the lidar SNR was fixed for all 2000 end-to-end retrievals. Lidar OC were
 301 fixed (albedo 0.3, VZA 0°, SZA 30°), but close to the median of those OC of the MCS, whilst the
 302 SZA is relatively low. This makes the solar background noise, the most dominant noise source, a
 303 conservative estimate.
 304



306 **Figure 2.** (a) Scheme of end-to-end simulation pipeline with geometry module (GM/OC), scene
 307 generation module (SGM), instrument module, L0-L1b module, and L1b-L2 module which
 308 includes the forward model. Example TOA and L1b spectra are shown. (b) Example discretized
 309 AOD profile as used in E2E. (c) Backscatter and attenuated backscatter profile input for lidar
 310 simulation. (d) Simulated range corrected lidar signal resulting from (c) with and without noise
 311 (SNR 4) and Gaussian fit.

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 313

314 4 Results

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318 4.1 Single central lidar with noise

318 For the first scenario, a single aerosol profile constrained by lidar soundings was assumed. Table
 319 2 shows the first 10 out of 2000 MCS results. Figure 3 shows the histograms of L2 and L3 results
 320 and L3 errors for the SRTM and NSRTM case. Also shown is the cumulative probability
 321 distribution of the absolute L3 errors for both models.

322 Not accounting for aerosol scattering in the RTM (using a NSRTM) leads to a negatively
 323 biased XCH₄ for all measurements (underestimated truth, Fig. 3a). Depending on the scenario
 324 (OC, aerosol profile), minimum L2 biases were -76 ppbCH₄ (-64 ppbCH₄) for plume (background)
 325 pixels (Fig. 3a). The mean absolute L3 bias magnitude is 2.56 ppbCH₄ (MAE, Fig. 3b). There are
 326 L3 bias magnitudes larger than that, associated with L2 bias below -10 ppbCH₄ (e.g., rows 2, 4,
 327 Table 2) where albedo was low (< 0.2) for plume and/or background. The corresponding
 328 mechanisms will be detailed in the discussions.

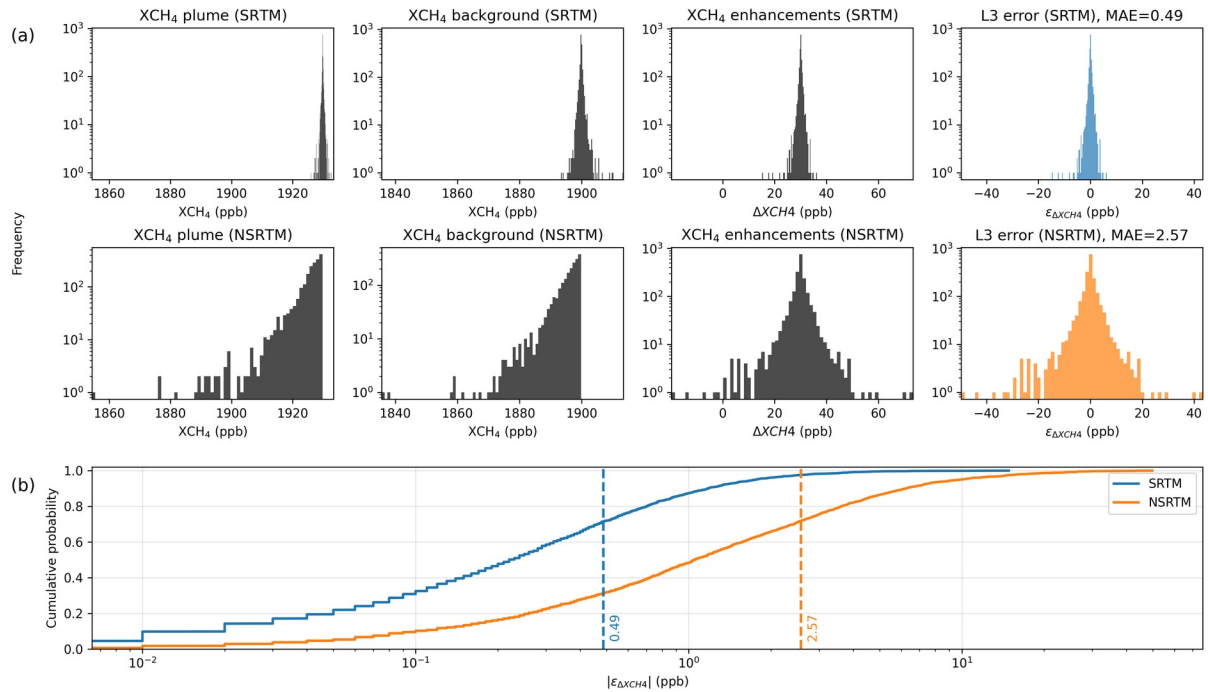
329 For the lidar-constrained SRTM, the shift of the cumulative probability distribution (Fig.
 330 3b) towards lower values, and the MAE of 0.49 ppbCH₄, indicates a substantial improvement of
 331 L3 bias averaged over the 2000 measurements. This is due to L2 biases being smaller than for
 332 the NSRTM and, due to lower unexplained residual radiance, more equal for plume and
 333 background, especially for measurements with higher plume/background albedo contrast (e.g.,
 334 rows 2, 5, 6 in Table 2). As further detailed in the discussion, the XCH₄ biases for the background
 335 are larger than for the plume, but still lower than for the NSRTM (spread of histogram, Fig. 3a).
 336

337 **Table 2. First 10 out of 2000 MCS results for the single central lidar with noise scenario. ret**
 338 **depicts retrieval, pl and bg stand for plume and background, respectively.**

ALH pl (m)	ALW pl (m)	AOD pl	ALH bg (m)	ALW bg (m)	AOD bg	ALH ret (m)	ALW ret (m)	Albedo pl	Albedo bg	SZA (°)	VZA pl (°)	VZA bg	VAA (°)	SAA (°)	XCH ₄ pl SRTM (ppb)	XCH ₄ bg SRTM (ppb)	XCH ₄ pl NSRTM (ppb)	XCH ₄ bg NSRTM (ppb)	$\epsilon_{\Delta XCH_4}$ SRTM	$\epsilon_{\Delta XCH_4}$ NSRTM
2208	1166	0.2	1950	1030	0.21	2211	1018	0.33	0.26	57	5	7	115	89	1929.85	1900.09	1927.47	1897.01	-0.24	0.46
2461	1342	0.29	2630	1440	0.3	2559	1351	0.14	0.44	51	17	20	138	185	1930.36	1899.66	1918.51	1895.74	0.7	-7.23
1005	832	0.18	780	640	0.19	956	1122	0.49	0.5	22	18	20	157	93	1929.9	1900.12	1928.8	1899.06	-0.22	-0.26
3648	875	0.42	4320	1040	0.39	3711	819	0.31	0.17	56	5	6	152	205	1929.75	1897.72	1918.54	1879.56	2.03	8.97
1245	1126	0.35	1180	1070	0.31	1242	846	0.27	0.1	24	10	13	23	166	1929.46	1900.07	1926.47	1893.46	-0.6	3.01
1911	820	0.27	1650	710	0.29	2044	1116	0.2	0.38	40	9	12	224	183	1930.36	1900.42	1924.69	1897.01	-0.06	-2.31
2852	993	0.3	2630	910	0.29	2851	972	0.47	0.32	35	6	7	110	126	1929.75	1900.29	1925.74	1894.5	-0.53	1.24
1628	832	0.32	1960	1000	0.35	1664	857	0.19	0.21	1	17	20	198	141	1930.3	1898.19	1921.43	1890.39	2.11	1.05
1759	758	0.02	2130	920	0.02	1731	898	0.43	0.54	19	7	9	55	131	1929.8	1899.77	1929.63	1899.64	0.02	-0.01
1864	494	0.02	1840	490	0.02	1970	481	0.26	0.41	47	6	8	81	128	1929.82	1899.82	1929.65	1899.71	0.00	-0.06

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340



341 **Figure 3.** Results for the single central lidar with noise scenario. (a) Histograms of plume and
 342 background L2 results, L3 results and L3 bias for the SRTM and NSRTM case. (b) Empirical
 343 cumulative distribution of absolute L3 errors for SRTM and NSRTM with MAE depicted.

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347 4.2 Fixed guess single central aerosol profile

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349 For the second scenario, the true aerosol profiles were equal to the previous scenario.
 350 However, no lidar aerosol profile measurements were used to constrain the SRTM. Instead, the
 351 retrieval ALH and ALW were fixed to 2000 m and 800 m, respectively, for all 2000
 352 measurements. For many of the 2000 measurements, the fixed guess aerosol profile was
 353 reasonably close to the true aerosol profile. For example, for almost half of the measurements,
 354 ALH was within 500 m of the truth. Hence, a L3 bias improvement below but comparable to the
 355 lidar scenario was achieved (MAE 0.82 ppbCH₄). This suggests that a SRTM constrained by a
 356 constant guess ALH and ALW, not necessarily from lidar, can be beneficial. Aerosol profile data
 357 could be obtained from external sources, such as EarthCare (Wandinger et al., 2023), TROPOMI
 358 (Sun et al., 2019) or the MODIS instrument (Lyapustin et al., 2011). Whether this could be done
 359 in near real time and close to the sounding area requires further investigation. Entirely relying
 360 on third party data sources could risk data availability, which would make a lidar preferable.
 361 Third sources could still be consulted to validate lidar data or to complement it, if lidar
 362 soundings fail.

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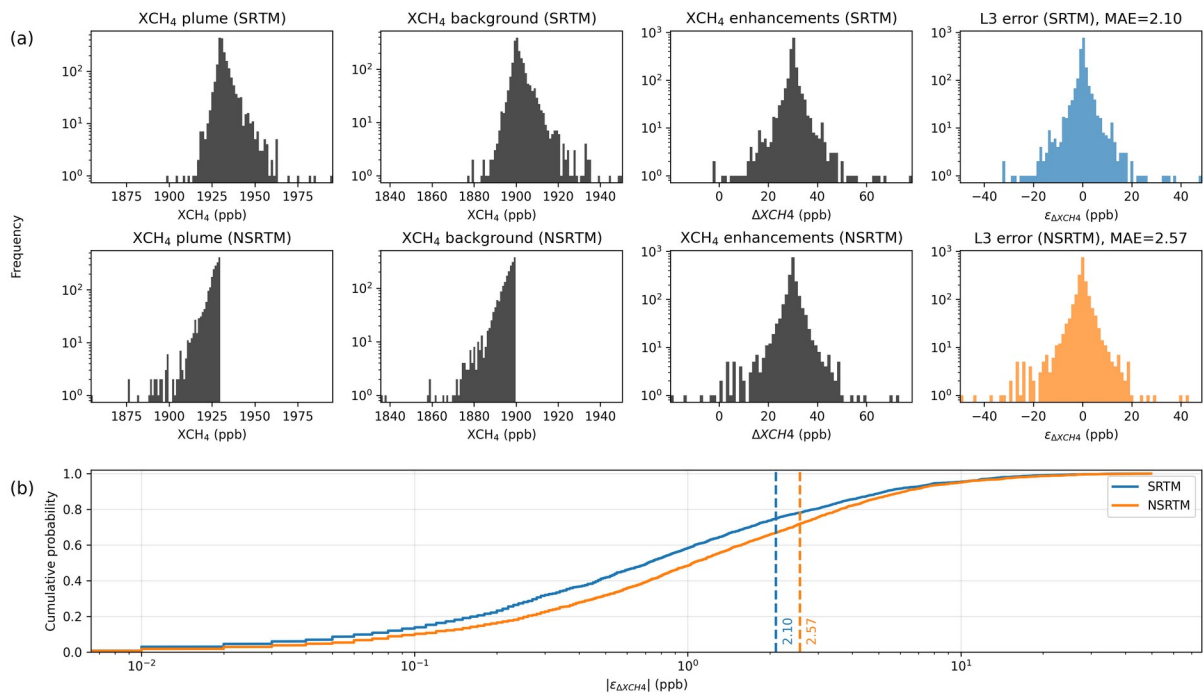
366

367 4.3 Random guess aerosol profile

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369 This MCS scenario was identical to the fixed guess run, except that for each measurement the
 370 SRTM was constrained with a random guess aerosol profile with mean/STD of 4000 m/3000 m
 371 for ALH and 1000 m/700 m for ALW.

372 The resulting MAE of 2.1 ppbCH₄ is comparable to ignoring aerosol extinction (NSRTM
 373 case). The L2 bias distributions are equally broad for SRTM and NSRTM (Fig. 4a) and cumulative
 374 probabilities are only slightly distinct (Fig. 4b). The overlapping tails indicate that frequency and
 375 magnitude of large L3 bias is not reduced. This suggests that a randomly constrained SRTM
 376 would not be beneficial for aerosol correction.



377 **Figure 4.** Results for the random guess scenario. (a) Histograms of plume and background L2
 378 results, L3 results and L3 bias for the SRTM and NSRTM case. (b) Cumulative distribution of
 379 absolute L3 errors for SRTM and NSRTM with MAE depicted.

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382 **5 Discussion**

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5.1 Level-3 bias and implications for methane fluxes (Level-4)

386 The MAE quantifies the absolute mean spread of the L3 (ΔXCH_4) bias values, $|\epsilon_{\Delta XCH_4}|$. For the
 387 single central lidar scenario, for both the NSRTM and SRTM, $\epsilon_{\Delta XCH_4}$ are fairly normal distributed
 388 with a mean close to zero for both NSRTM (-0.27 ppbCH₄) and SRTM (-0.02 ppbCH₄). The SRTM,
 389 if sufficiently constrained, thus reduces the magnitude of the L3 bias on average, but it does not
 390 cause a notable systematic shift in ΔXCH_4 .

391 The transported mineral dust aerosol scenario entailed a MAE of ~ 2.6 ppbCH₄, when
392 ignoring aerosol extinction in the retrieval (NSRTM in Table 3). L3 bias ranged from -49.8
393 ppbCH₄ to 43.3 ppbCH₄ (Fig. 3a). The SRTM constrained by a single aerosol profile from only the
394 plume, but not the background, partly corrects for scattering at both plume and background. It
395 increases accuracy of both plume and background forward modeled radiance, and
396 consequently reduces XCH₄ bias for both. This leads to a reduced L3 bias magnitude (Eq. (4)) on
397 average, i.e., a reduced MAE. With lidar, or a close guess, the MAE was reduced to 0.49 ppbCH₄
398 and 0.83 ppbCH₄, respectively (scenarios 1 and 2, Table 3).

399 What would be the L4 (flux) error when ignoring aerosol scattering? Assuming a
400 constant column averaged ΔXCH_4 of 30 ppbCH₄ across the plume, the MAE of 2.6 ppbCH₄
401 corresponds to an average relative L3 error of $\pm 8.5\%$. Adopting the aforementioned plume
402 model (160 m wide plume, 1 ms⁻¹ wind speed, ~ 100 kgCH₄h⁻¹), to first order, the propagated
403 average bias would be roughly ± 9 kgCH₄h⁻¹. MCS were repeated for column enhancements of
404 20 , 10 and 5 ppbCH₄. The bias remained approximately constant. This suggests that for lower
405 CH₄ fluxes, the average relative L3 (and L4) error would be higher, e.g., $\sim 27\%$ for a ΔXCH_4 of 10
406 ppb, provided enough sensitivity to resolve this enhancement.

407 Even if aerosol scattering is constrained, there are few cases with considerable L3 bias
408 (Fig. 3b). However, these cases are significantly reduced in magnitude and occurrence
409 compared to an unconstrained SRTM or a NSRTM. This is reflected in the tails of the L3 error
410 distribution. In the lidar constrained SRTM scenario (Fig. 3b), for only 47 cases ($\sim 2\%$) the L3 bias
411 magnitude was > 2.6 ppbCH₄. For 8 measurements (0.4% of all measurements), the L3 bias
412 magnitude was ≥ 5 ppbCH₄, the highest corresponding to -14.85 ppbCH₄. These are cases with a
413 large difference between retrieval ALH and background ALH, which the lidar does not measure.
414 The underlying mechanisms will be detailed further below.

415 For those cases, however, the NSRTM L3 bias was even higher. For the NSRTM, 556
416 measurements ($\sim 28\%$) entail a L3 error magnitude > 2.6 ppbCH₄. In 270 cases ($\sim 13.5\%$ of
417 measurements) the L3 bias magnitude was ≥ 5 ppbCH₄, with extreme values of -49.8
418 ppbCH₄/ 43.3 ppbCH₄ (min/max), that is, higher than the 30 ppbCH₄ enhancement. Therefore, in
419 13.5% of the cases, L3 bias would be $\sim 17\%$ to $> 100\%$, with corresponding first order L4 errors.

420 The L3 biases of this study are in line with those found by Yu et al. (2024), who
421 simulated enhancement methane bias near 1666 nm for different aerosol types near industrial
422 sites and with the lower end L3 biases found by Huang et al. (2020), who simulated L3 bias near
423 2 μ m for an airborne sensor altitude using an aerosol mixture.

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436 **Table 3.** Summary of all MAE for all simulated scenarios.

Nr.	Scenario	MAE (ppbCH ₄) (SRTM)	MAE (ppbCH ₄) (NSRTM)
1	Single central lidar with noise	0.49	2.57
2	Constant guess aerosol profile	0.83	2.57
3	Random guess aerosol profile	2.10	2.57
4	Single central lidar with noise, aerosol near ground	0.25	1.29
5	Ideally constrained, different aerosol profile, equal OC	0.003	0.69
6	Ideally constrained, constant aerosol profile, equal OC	0.002	0.09
7	Ideally constrained, constant aerosol profile, different OC	0.003	2.50

437

438

439 5.2 Level 2 bias

440

441 The observed mean L2 error over all 2000 runs, when not accounting for aerosol scattering
 442 (NSRTM), is -5 ppbCH₄ (-4.7 ppbCH₄) for the plume (background), or about -0.26%, and thus
 443 compatible to Butz et al. (2012), who simulated aerosol induced L2 bias in the 2 μm spectral
 444 region.

445

446 5.2.1 Bias mechanisms for the NSRTM L2 bias

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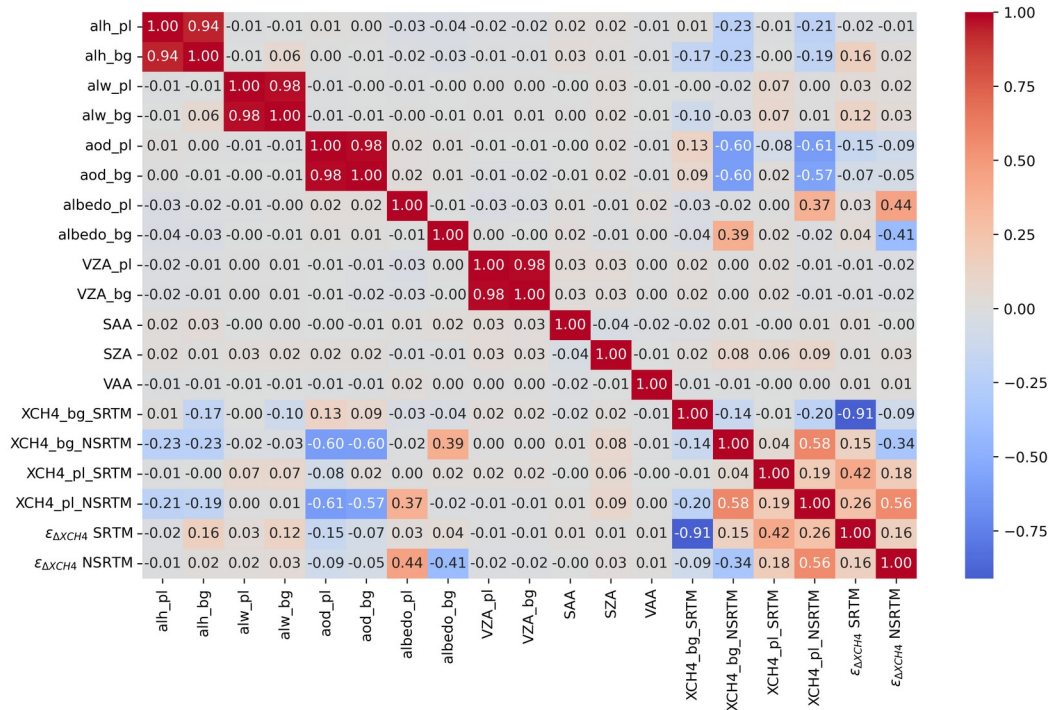
448 The negative bias as a result of ignoring aerosol extinction in L2 retrieval can be explained by a
 449 path shortening effect, which is pronounced when the aerosol layer is located above the
 450 methane accumulation in the lower atmospheric boundary layer. This reduces the sensitivity of
 451 the simulated TOA radiance to the near-surface, methane-rich atmospheric layers. The
 452 apparent depth of the molecular absorption lines is smaller, similar to a dimming effect, since
 453 less photons traverse the near-surface layers. Once albedo cannot be optimized further, the
 454 NSRTM retrieval lowers the trace gas vertical column density (VCD), resulting in a systematic
 455 underestimation of absorption and thus total column XCH₄. This is also reflected in the anti-
 456 correlation between XCH₄ and AOD and the weak anti-correlation of XCH₄ with ALH (Fig. 5): The
 457 higher the aerosol layer, the stronger the path shortening effect, the larger the L2 bias
 458 magnitude (e.g., row 3 vs. 7, Table 2). Path shortening also applies to XCO₂, which is co-
 459 retrieved in the retrieval with 425 ppm ground truth. The bias is -1.2 ppm XCO₂ on average,
 460 with extreme values of -18.3 ppm (plume) and -15.8 ppm (background), demonstrating the
 461 working principle of the CO₂ proxy method. XCO₂ from SCIAMACHY was indeed found to vary by
 462 up to 37 ppm over the Sahara due to mineral dust aerosol (Houweling et al., 2005).

463 Figure 5 furthermore shows a moderate correlation between albedo and XCH_4 for both
464 plume and background pixels. A higher albedo indicates a stronger contribution of direct
465 surface reflected radiance relative to the scattered radiance (surface coupling). For higher
466 surface coupling, path shortening is less prominent and the L2 bias magnitude is smaller
467 compared to measurements with lower albedo. This correlation is really associated with an
468 interplay between surface coupling and path shortening. For example, the path shortening
469 mechanism leads to particularly strong L2 bias when, additionally to a high ALH (clearly above
470 the peak methane concentration, >1500 m), the albedo is low (<0.2, e.g. rows 2, 4, 5 in Table 3).

471 For lower ALH, therefore, the aerosol would be located nearer to the maximum trace
472 gas retrieval sensitivity near the ground, and path shortening would be reduced. This was
473 confirmed with a separate MCS using a mean/STD for ALH of 1000 m/700 m, while mean/STD
474 of ALW remained at 700 m/400 m. The MAE decreased to 1.29 ppb CH_4 and 0.25 ppb CH_4 for the
475 lidar-constrained SRTM and NSRTM, respectively (scenario 4, Table 3).

476 Since the single scattering simulation scheme used here does not model multiple
477 scattering, which would also allow path lengthening and thus a positively biased XCH_4 , the true
478 XCH_4 forms a hard upper limit and the distribution appears truncated (Figs. 3a). The degree of
479 truncation depends on the measurement altitude. When the latter was moved downward, e.g.,
480 at 5 km above ground level, practically simulating an airborne geometry, the likelihood of
481 aerosol layers being located above the sensor increased. For these cases, the scattered radiance
482 contribution decreased, while surface coupling increased and positive bias became more likely,
483 especially for high albedo (>0.3), leading to a more symmetrical L2 distribution (as already
484 visible in Fig. 4b).

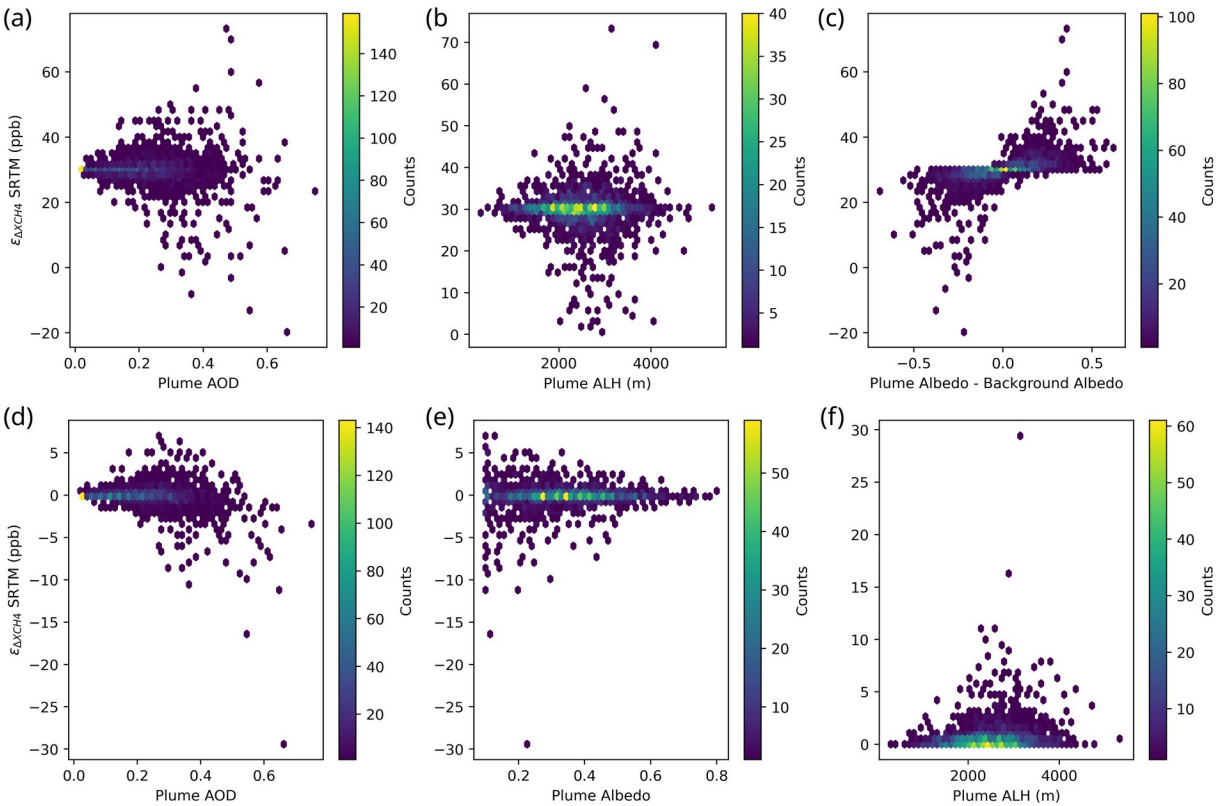
485 As they are drawn from a normal distribution, OC are roughly equally likely for plume
486 and background and therefore positive and negative L2 biases have equal likelihood too. As a
487 result, ΔXCH_4 and L3 bias distribution are symmetric (Figs. 3a, 4a).



488 **Figure 5.** Pearson cross correlation matrix for the MCS parameters for the lidar scenario
 489 (section 4.1). There are moderate correlations between independently calculated variables, and
 490 thus are non-causal, such as between plume pixel albedo and background XCH₄. Since the L3
 491 error is the difference between the retrieved and the true L3 enhancement (Eq. (4)), which is
 492 fixed at 30 ppbCH₄, the L3 bias correlates (anti-correlates) with the plume (background) XCH₄
 493 for both NSRTM and SRTM. Weak correlation values (<0.4) may indicate multi-parameter
 494 dependence, which is not fully captured by pairwise Pearson correlations.

495
 496 A strong L2 bias, e.g., due to high ALH, and/or low albedo and thus low surface coupled
 497 radiance, alone does not cause a large L3 bias magnitude, if plume and background albedo are
 498 comparable (within ~50%) and/or AOD is low (e.g., rows 3, 9, 10 in Table 2). If, however, due to
 499 different OC, path-shortening and surface coupling work disproportionately for plume and
 500 background, the L2 bias inequality increases and, as a result, the L3 bias magnitude increases
 501 (Eq. (4)). This effect is promoted by high AOD and ALH (Figs. 6a and b), i.e., strong path-
 502 shortening. Due to the assumption of a smoothly varying aerosol layer, plume and background
 503 ALW, AOD and ALH strongly correlate, and plume/background albedo contrast is the strongest
 504 driver of L2 bias inequality and thus L3 bias (Fig. 6c), while VZA differences have little impact
 505 and VAA per default are equal for plume and background. For instance, if the plume pixel
 506 albedo is lower than the background albedo, plume XCH₄ is more negatively biased than
 507 background XCH₄, and, with Eq. (2), L3 bias is negative (e.g., row 2 in Table 2). A positive L3 bias
 508 would be associated with a plume surface albedo larger than background albedo (e.g., rows 4, 5

509 in Table 2). Therefore, differences in OC drive the L3 bias magnitude, which relates to the core
 510 hypothesis.



512 **Figure 6.** Scatter plots color coded for occurrence showing L3 bias against different MCS
 513 parameters. (a) to (c): for the lidar scenario for NSRTM (scenario 1, Table 3) (a) AOD. (b)
 514 plume/background albedo contrast. (c) ALH. (d) to (f): Scatter plot for the NSRTM case for
 515 scenario 5 in Table 3 (with equal albedo for plume and background). (a) AOD, (b) albedo (c)
 516 ALH. Only plume parameters are shown, since plume and background aerosol parameters are
 517 strongly correlated.

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519

520 As will be investigated in more detail in the following, L2 bias inequality occurs even for
 521 equal OC and reasonably similar aerosol profiles, but is amplified by difference in aerosol profile
 522 parameters, and more so, OC differences between plume and background. To further isolate
 523 governing mechanisms and provide further testing evidence for the null-hypothesis, three
 524 reduced complexity tests were carried out. The SRTM was ideally constrained for all three. This
 525 means, while still random, the retrieval aerosol parameters (ALH, ALW, AOD) were equal
 526 to the true values for both plume and background.

527

528 For the first reduced complexity scenario, OC (albedo, VZA, VAA) were identical for
 529 plume and background. As the SRTM was constrained with the true aerosol profiles, the
 530 corresponding MAE is ~ 0 (scenario 5, Table 3). The L2 bias distribution is nearly delta-like for
 531 both plume and background, and L3 bias is ~ 0 for all measurements. For the NSRTM, the MAE
 reduced from 2.57 ppbCH₄ to 0.69 ppbCH₄, with L3 bias ranging between about -11 and 4

532 ppbCH₄. Despite equal OC, the non-zero MAE of 0.69 ppbCH₄ implies that a number of
 533 measurements entailed cases with a plume/background L2 bias inequality, caused by
 534 plume/background difference in aerosol profile. For these measurements, albedo was low
 535 (<0.2) and/or AOD was high (Figs. 6d and e), i.e., radiance surface coupling was weak. This
 536 increased the sensitivity of the retrieval to aerosol-induced path shortening, causing small
 537 differences in effective optical path between plume and background (e.g. different ALH) to map
 538 into disproportionate L2 biases and thus higher L3 bias. As before, XCH₄ and ALH anticorrelated
 539 for plume and background, meaning large ALH and large ALH differences promoted higher L3
 540 bias (Fig. 6f). No notable dependence on ALW was found.

541 The second reduced complexity scenario (scenario 6, Table 3) was identical to the first
 542 (equal OC, true aerosol profile for retrieval), except that differences in aerosol profile were
 543 removed. The same aerosol profile was used for plume and background. The MAE for the SRTM
 544 was ~0 again, as expected. For the NSRTM, min/max L2 bias was -31 ppbCH₄/-0.3 ppbCH₄
 545 (plume) and -30.4 ppbCH₄/-0.3 ppbCH₄ (background) and the MAE was 0.09 ppbCH₄. This small
 546 MAE confirms the trivial case of canceling L2 bias (Eq. (3)) due to equal aerosol profiles for
 547 plume and background, if OC are the same. Methane does not contribute to scattering.
 548 However, differences in methane VCD between plume and background modify the attenuation
 549 of scattered photons (Eq. 6 in supporting information), leading to plume/background
 550 differences in the relative contribution of scattered radiance to the total signal. This explains
 551 small L2 bias inequalities and thus non-zero (sub-ppb) L3 bias even in this case.

552 The third run was identical to the second run (constant aerosol), but OC differed again
 553 (scenario 7, Table 3). MAE was 2.5 ppbCH₄, i.e., similar to the MAE with a varying aerosol profile
 554 (e.g. scenario 1, Table 3). This indicates that aerosol profile differences do indeed amplify L3
 555 bias, but less so than differences in OC, which in this study practically means albedo.

556 In summary, the reduced complexity tests show that for identical aerosol profiles for
 557 plume and background and OC (mainly albedo), the radiative transfer is more similar for both.
 558 As a consequence, the associated L2 biases approximately cancel, leading to a low L3 bias
 559 magnitude. As the aerosol profile varies between plume and background, differences in path
 560 length, scattering phase function and radiance surface coupling increase, and cause noticeable
 561 L3 biases for most measurements. These effects are further enhanced once OC between plume
 562 and background differ. The assumption of canceling L2 biases (Eqs. (2) and (3)) does no longer
 563 hold as soon as OC differ, regardless if the aerosol profile is constant or varying between plume
 564 and background.

565 Overall, the result suggest that a constrained SRTM becomes more useful the more OC,
 566 most importantly albedo, differ between plume and background. This is likely the case, given
 567 the GSD of least 50 m, plume dimensions of hundreds of meters and a lidar footprint of ~130 m
 568 (Fig. 1b)

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571 5.2.2 Bias mechanisms for the SRTM

572

573 As noted above, 0.4% of lidar-constrained measurements still exhibit L2 bias magnitudes ≥ 5
 574 ppbCH₄, with plume (background) L2 biases ranging from -7 to 13 ppbCH₄ (-4 to 3 ppbCH₄).

575 These are associated with large differences between retrieval and background aerosol profiles.

576 The same principal mechanisms that govern L3 bias for the NSRTM (surface coupling vs.
577 path shortening) are at play, but they become pronounced more the poorer the SRTM is
578 constrained. In the physics-based retrieval framework used here, the aerosol vertical profile
579 (AOD, ALH, ALW) is fixed, which may lead to path-length mismatches. Trace gas VCD and
580 surface albedo are the only adjustable state-vector elements. Hence, L2 bias, and therefore L3
581 bias, are partly driven by how close retrieval and true aerosol profiles are. For instance, if the
582 retrieval ALH is higher than the true ALH (measured spectra with deeper methane absorption
583 features than explainable with fixed aerosol profile), forward modeled path shortening is
584 overestimated, which is compensated by increasing model VCD, causing positive XCH₄ bias.
585 AOD mismatch also contributes and is more pronounced for higher AOD. This mechanism likely
586 contributed to a slight asymmetry around the true XCH₄ for both plume and background (Fig.
587 4a), with large positive L2 bias occurring more frequently than large negative L2 bias. The larger
588 errors of aerosol profile parameters explain the higher occurrence of larger XCH₄ biases for the
589 background and thus the broader distribution (SRTM, Fig. 3a).

590 A loosely constrained SRTM, therefore, performs equally poor as a NSRTM, such as
591 shown with the random guess scenario (Fig. 4). For both NSRTM and SRTM, relatively large L2
592 biases of tens of ppbCH₄ occur, with values between about -32 and 50 ppbCH₄ (Fig. 4a). L2 bias
593 approximately canceled when the retrieval aerosol profile was relatively close to the true
594 aerosol profile, but led to large L3 bias otherwise, particularly in combination with high
595 plume/background albedo differences. Largely overestimated ALH in the SRTM caused
596 overestimated path shortening, corresponding to an AK with misallocated sensitivity, leading to
597 overestimated (positively biased) XCH₄, which was amplified for high plume/background albedo
598 contrast. On the contrary, for measurements with stronger surface coupling (albedo >0.2, AOD
599 <0.2), aerosol scattering less dominated the TOA radiance and aerosol profile errors had a
600 lower impact on the averaging kernel. Depending on the OC, we found that ALH accuracy
601 should be a few hundred meters in order for the L3 bias magnitude to remain <10 ppbCH₄ (<
602 30% of enhancement).

603

604

5.4 Other limitations of the simulation scheme

605

606 The relationship between the discussed aerosol-induced mechanisms and the L2 and L3 biases
607 depend to some extent on the retrieval model and its limitations. For example, Huang et al.
608 (2020) found that, depending on the retrieval model used, the L3 bias was either independent
609 on XCH₄ or it decreased with increasing XCH₄. Here, we found that L2 bias remained almost
610 constant (0.05 ppb decrease for 10 ppb reduction in true Δ XCH₄) and L3 bias would remain ~2.6
611 ppbCH₄ even with lower CH₄ enhancements.

612 The forward modeling scheme used here was able to simulate a limited ensemble of
613 processes only. For instance, multiple scattering was not modeled. Although representative,
614 only a single aerosol type has been considered. The two-pixel assumption is another major
615 simplification, which, however, allowed to focus on fundamental mechanisms to assess the
616 principle suitability of a aerosol profile from lidar to constrain the retrieval model. However,
617 even full-physics retrievals are limited (Frankenberg et al., 2025). In line with this study, more
618 complex simulations of surface and atmospheric scattering also found that albedo induced bias
619 is amplified with aerosol abundance, as observed in XCH₄ from TROPOMI (Somkuti et al., 2025).

620 Being sampled independently, the MCS entailed less likely combinations of aerosol
621 parameters, such as AOD and ALW. However, these represent values from the tails of the
622 distribution. While they would not alter the main results, they helped to unravel the
623 mechanism of the model. For example, an AOD of 0.02 and ALW of 758 m (row 9, Table 2)
624 would correspond to practically clear air, rather than the intended dust aerosol scenario.
625 Further, the lidar would neither be measuring any signal nor be needed in that case, consistent
626 with the discussion above.

627 The finding that albedo difference is the dominating driver of L3 bias over aerosol profile
628 differences is based on a smoothly varying layer (mean difference 205 m/62 m/0.02 for ALH,
629 ALW and AOD). For strongly varying aerosol layers, such as a cloud edge, large L3 bias would
630 arise even at equal albedo. This has been assessed in a dedicated simulation with a stratus
631 cloud scenario.

632 Due to computational constraints, the vertical grid spacing was 1000 m, as in a real data
633 processing. This led to under-sampling of some aerosol ALW, thus contributing to a flattening of
634 the relationship with ALW. However, repeating the lidar MCS using 250 m grid spacing yielded
635 the same MAE and differences in XCH_4 were $<0.05\%$.

636

637 **6 Conclusions**

638 The potential to constrain a physics based methane retrieval scheme for space-borne imaging
639 spectroscopy with aerosol layer height and width (ALH, ALW) from lidar was assessed. The
640 enhancement (ΔXCH_4 , plume) is retrieved as the difference between the XCH_4 of the plume
641 pixels and the XCH_4 at the background pixels next to the plume. The hypothesis was tested that
642 knowledge of the aerosol profile from a single staring lidar, probing the plume only (assumed to
643 be at swath center), would not be beneficial to reduce the bias of the methane Level-3 (L3)
644 results ΔXCH_4 , since it would miss aerosol profile at the background area of a given scene. In
645 the case of a constant aerosol profile for plume and background, the Level-2 (L2) biases of
646 plume and background would possibly cancel, leading to nil L3 bias and a lidar would be
647 redundant.

648 To test the hypothesis, end-to-end processing was performed in Monte Carlo simulation
649 (MCS) mode with 2000 retrievals from simulated TOA spectra up to L2 (XCH_4) and L3 (ΔXCH_4)
650 using a non-scattering RTM (NSRTM, ignoring aerosol extinction) and a scattering RTM (SRTM,
651 accounting for aerosol extinction assuming single scattering). A transported mineral dust layer
652 was assumed, covering the complete scene, smoothly transitioning between plume and
653 background, represented by a single pixel each. Based on the results, the following main
654 conclusions are drawn:

- 655 (a) For equal OC, notably albedo, and constant aerosol profile between plume and
656 background, the L2 bias for plume/background is approximately equal and cancels,
657 leading to small L3 bias in most cases ($\ll 1$ ppb CH_4), even without correcting for aerosol
658 (scenario 6, Table 3).
- 659 (b) For different OC, significant L2 bias inequalities may arise, caused by a combination of
660 path-shortening and surface coupling, with L3 bias values of several tens of ppb CH_4 for a

- 661 NSRTM or a poorly constrained SRTM, regardless if the aerosol profile varies between
 662 plume and background or not.
- 663 (c) Therefore, plume/background L2 bias does generally not cancel if plume/background
 664 OC, notably albedo, differ.
- 665 (d) A (lidar-) constrained SRTM is effective for high albedo contrast, even for a smooth to
 666 constant aerosol profile, such as an extended mineral dust layer; and even for
 667 homogeneous albedo, if albedo is low, AOD and ALH are high and plume/background
 668 aerosol variations are high (Table 3).
- 669 (e) In the MCS, a lidar-constraint SRTM with AOD uncertainty <10% reduced the L2 bias and
 670 L2 bias inequality and thus L3 bias for most of the 2000 measurements (L3 MAE ~0.5
 671 ppbCH₄ with min/max L3 bias of -14.9/6.3 ppbCH₄). The occurrence of large L3 bias (≥5
 672 ppbCH₄ or ~17% of enhancement), which, to first order, would propagate to a L4 (flux)
 673 error of ≥17%, is reduced from 13.5% to 0.4%. This refutes the null hypothesis.
- 674 (f) ALH accuracy must be a few hundred meters for L3 bias magnitudes to remain <10
 675 ppbCH₄ (i.e., <30% of ΔXCH₄)
- 676 (g) AOD needs to be known better than 50% for a lidar-constraint SRTM to be efficient
 677 (supporting information, text S2)
- 678 (h) Third party aerosol profile data may complement lidar retrievals, for instance, if lidar
 679 soundings fail.

680

681 If AOD cannot be inverted from lidar data, it could be retrieved from observational networks
 682 such as AERONET (Holben et al., 1998) or L2 data from EarthCare (Wandinger et al., 2023). For
 683 further reaching conclusions, a more detailed assessment on spatio-temporal coverage and
 684 availability and accuracy of such data is needed.

685 In the model used here, MAE was found to vary very little with ΔXCH₄. Since L4 bias to
 686 first order scales with the L3 bias, this suggests, therefore, that aerosol bias correction with
 687 lidar data would be particularly beneficial for smaller ΔXCH₄ and hence smaller fluxes (<100
 688 kgCH₄h⁻¹) or higher diluted plumes. More leaks could be detected and more plumes could be
 689 quantified. In the US oil and gas sector ~70% of the methane sources emit less than 100 kgCH₄h⁻¹
 690 ¹ (Williams et al., 2025). Of course, this is provided that the sensitivity of the instrument-
 691 retrieval system is sufficient to resolve those smaller enhancements.

692 In summary, the study suggests that, given an extended mineral dust aerosol layer, a
 693 single lidar profile is beneficial for space-borne methane plume quantification, especially for
 694 large albedo contrast between plume and background scene pixels, as they may arise in reality
 695 (Fig. 1). The result informs next steps, which will include high altitude measurements to validate
 696 the benefit of the aerosol lidar in methane remote sensing.

697 Since CO₂ is co-retrieved and proportionally biased by aerosol, CO₂ plume quantification
 698 would equally benefit especially in areas where other aerosol-correction methods, such as the
 699 proxy method, fail. There are implications beyond man-made methane or CO₂ emissions. For
 700 instance, volcanic aerosols, including sulfur species, are a major constituent of magmatic
 701 degassing of CO₂. A lidar constrained retrieval could allow retrieving more accurate CO₂ fluxes,
 702 and in combination with a full physics retrieval, possibly a more accurate retrieval of volcanic
 703 aerosol properties, such as aerosol layer height (Theys et al., 2022). This could allow a more
 704 accurate quantification of the geological carbon cycle (Werner et al., 2019).

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706

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713

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715 **Data Availability Statement**

716 The E2E is based on the SRON open source Toolbox for End-to-End Simulations, which can be
717 downloaded from

718 <https://teds.readthedocs.io/en/latest>

719 The aerosol type specific single scattering albedo and scattering phase function was taken from
720 the OPAC catalogue (<https://geisa.aeris-data.fr/opac/>). ALH, ALW and AOD were adopted from
721 the AERONET observational network (<https://aeronet.gsfc.nasa.gov/>). All results (~ 51 GB of
722 data) are available upon request.

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725 **Conflict of Interest Disclosure**

726 The authors declare there are no conflicts of interest for this manuscript

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728 **References**

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