Response to Interactive Comment from Referee #2

General Comment

This paper presents vertical profiles of aerosols, NO2, and SO2 retrieved from MAXDOAS measurements at the Fort McKay South field site (Alberta, Canada). This site is located close two mining plants which are major sources of industrial pollution in this region. The MAX-DOAS retrieval results are compared to co-located ancillary observations from lidar, AERONET, active DOAS, Pandora, and airborne in-situ analysers instruments. These comparisons based on data sets from various techniques provide a unique opportunity to investigate the performance of MAX-DOAS retrievals under varying atmospheric conditions in an industrial area.

The manuscript is well written and clearly structured, and presents very interesting results which fit well with the scope of ACP. I recommend the final publication of the manuscript after addressing the following specific comments and technical corrections:

Response: The authors thank referee #2 for reading and commenting on the discussion paper. We are pleased that it meets with your approval, pending the specific comments and technical corrections below.

Specific Comments

1. Page 6, lines 1-12: It is not clear in which direction the active-DOAS measurements are performed. Is it the same direction as the MAX-DOAS instrument ? Maybe this information could be added in Figure 1.

Response: Direction of active-DOAS light path added to Page 6 lines 2-3.

2. End of page 7-beginning of page 8: You should add Wagner et al., Atmos. Meas. Tech. (2019) in the list of references on the O4 scaling factor. No O4 scaling is used in the present study. Did you perform sensitivity tests on your aerosol retrievals and you came to the conclusion that a scaling factor was not needed ? Or you simply decided not to use any scaling factor ? I think this should be further discussed in the paper and sensitivity test results could be also added to make the study more robust (e.g. what is the impact of a scaling factor on the agreement with AERONET data ?).

Response: The Wagner et al. 2019 reference was added on page 8 line 5. A statement that the scaling factor was not used because of a lack of conclusive need based on the literature and the good agreement between the lidar and MAX-DOAS AODs when the modelled S-ratios were applied on Aug 23 was added on page 8 lines 7-9.

Note that while the lidar and MAX-DOAS AODs are expected to be equal within error because the 0-4 km lidar profiles of aerosol extinction were smoothed with the MAX-DOAS retrieval

information, the AERONET AODs are expected to be greater than both because the sun photometer observed the entire atmospheric column. Since the MAX-DOAS AODs retrievals used an exponentially decreasing a-priori that reduced to near-zero (<0.005) extinction above 2 km (chosen for reasons discussed in Supplemental Section 8.2.2), the smoothed lidar and MAX-DOAS AODs are expected to capture the extinction between 0-2 km. The AERONET AODs would only be expected to be the same as the MAX-DOAS AODs if either a) there was no aerosol extinction above the boundary layer or b) the AERONET AODs were converted to a 0-4 km profile that was smoothed using the MAX-DOAS retrieval information. There was evidence of non-trivial aerosol extinction above 2 km and even sometimes above 4 km in the lidar measurements. An example of the variation in aerosol extinction at altitudes above 2km observed by the lidar was added to the supplemental (Fig. S9). An expanded discussion of how contributions of aerosol extinction above the boundary layer to the total AOD can be non-trivial was added on page 14 starting on line 5. This includes a reference to measurements of monthly average contribution of free tropospheric AOD to total AOD from satellite observations (Bourgeois et al., 2018, Atmos. Chem. Phys. DOI: https://www.atmos-chem-phys.net/18/7709/2018/).

3. Page 9, line 3: According to Rodgers (2000), weighting function K should be equal to $\delta y/\delta x$ and not $\delta F/\delta x$.

Response: corrected on Page 9 line 8.

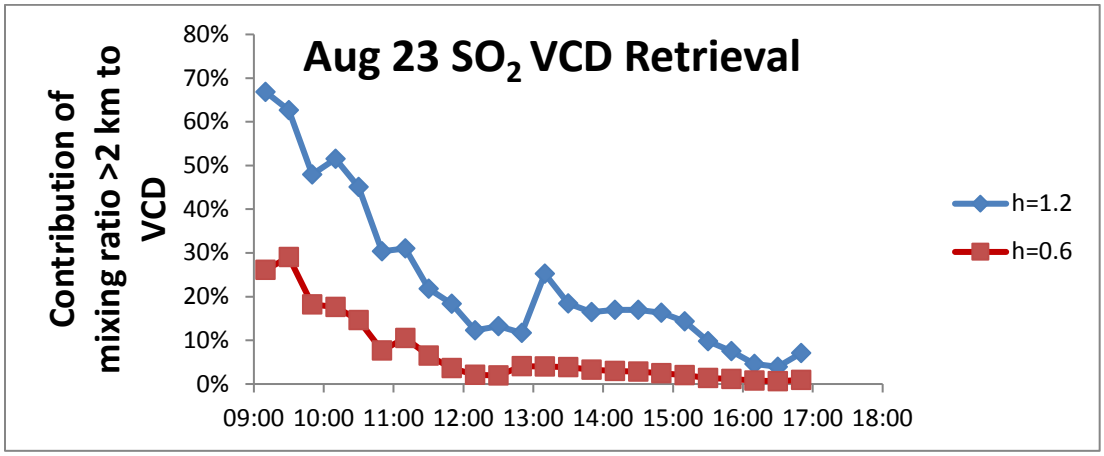
4. Page 9: I think you should justify your choice of aerosol extinction and trace gas concentration a priori profiles. Did you perform sensitivity tests for the selection of these a priori profiles, especially in terms of scaling height ?

Response: Results and discussion of sensitivity analysis performed on the a-priori profiles was added to the Supplemental as Section 8.2. Sensitivity studies are shown for two case study days, Aug 23 and Sep 04, including results of a-priori scaling height of 0.3, 0.6 and 1.2 km. The new section is now referred to in the main manuscript on pages 9 line 22 and 10 line 2.

Regarding variations in the scale height (except from discussion in supplement 8.2): The a-priori with a scale height of 0.3 km decreased the AODs or VCD retrievals by 0-30% compared to the "base-case", depending on the day and species (Tables S18-S23). The retrievals using this a-priori resulted in greater chi-squared values for the h= 0.3 profiles compared to the other profiles for some retrievals after 15:00 (Figs. S14-S16). This smaller a-priori scale height was not used for the retrievals because it was probably too restrictive in terms of the decrease in the a-priori values profile values between ~0.5-1 km under well-mixed, afternoon boundary layer conditions.

The a-priori with a scale height of 1.2 km increased the AOD or VCD retrieved values by ~20% compared to the "base-case", generally because there was little information content from the MAX-DOAS measurements above ~1.5 km and the retrieval reverted to the non-zero a-priori values (Figs. S21-S25). Consequently, this profile shape increased the proportion of aerosol

extinction and trace-gas concentration present at >2 km altitude to the 0-4 km column value (see plot below and Figs. S21-S25). For some of the vertical profiles on both days, the retrieval reduced the aerosol extinction to zero by ~1 km but this a-priori resulted in non-zero values at 2-4 km (Figs. S21-S25). This larger scale height was not used for the retrievals because it resulted in non-zero values at high altitudes where there was little to no information content from the measurements (Figs. S7, S21-S25). Since the values at these higher altitudes were unknown and likely varied temporally, a scale height that allowed the values to be retrieved at zero was chosen (i.e., h= 0.6 km). An a-priori scale height that resulted in underestimated rather than overestimated retrievals of the pollution loading where there was a lack of information content was considered preferable.



5. Page 9, lines 30-31: To my knowledge the SCIATRAN RTM is not based on a Monte Carlo approach. This point should be clarified.

Response: The reviewer is correct. "Monte Carlo" was removed on page 10 line 8.

6. Page 10, line 10: The relative error of the a priori was set to 100% for the construction of the Sa matrix. Did you set the extra-diagonal terms to zero and, again did you perform sensitivity tests for the selection of this relative error value. Also related: nothing is said in the paper about the quality control of your MAX-DOAS retrievals. For instance, what are the typical degrees of freedom for signal (DOFS) values of your aerosol and trace gas MAX-DOAS retrievals and what is the level of agreement between measured DSCDs and those modelled using the retrieved profiles as input ?

Response: Extra-diagonal terms of S_a matrix were set to zero and this information was added on page 10 line 19. Sensitivity tests for this relative error value were not performed. Typical degrees of freedom of signal were 1.6-2.1 and 2.3-3.0 for the aerosol and trace-gas retrievals, respectively. This information was added to Supplemental Section 8. Statistical results of the linear correlations between the measured and modelled dSCDs of O_4, SO_2, and NO_2 that includes

a brief discussion of results were added to the supplemental (section 8.1 Tables S10 and S11). The presence of a "quality of retrieval" section the supplemental was added on page 8 lines 29-30 of the manuscript.

7. Comparisons between MAX-DOAS and AERONET AODs (Figs 4a-9a): MAX-DOAS significantly underestimates (by sometimes more than a factor of 2) AERONET AODs. What happens if you used a different a priori profile with a scaling height larger 0.6 km (e.g. 1.2 km) ? Does it improve the agreement with AERONET without degrading the quality of your retrieval (see point 6/) ? Also, is the application of a O4 scaling factor can improve the agreement with AERONET ?

Response: See response to reviewer comment #2 for discussion of why MAX-DOAS AODs are expected to be significantly less than the AERONET AODs during certain periods such as on Sep 04. As now shown in Supplemental section 8.2, increasing the scaling height of the MAX-DOAS AOD retrieval to 1.2 km from 0.6 km for Sep 04 increased the diurnal AOD by 14% (+/- 13%) (Table S21). The AODs produced by the two scale heights were effectively equal within error for this day (Fig. S17). This result was expected since there appeared to be a significant contribution of aerosol extinction above the boundary layer height on this day that probably occurred on other days as well (see response to comment #2). The Aug 23 MAX-DOAS AODs using a scale height of 1.2 km were significantly greater than the h=0.6 km AODs between 15:30 and 16:45 but were significantly smaller after 17:00 with reduced degrees of freedom of signal (Fig. S14). The h=1.2 km AOD retrieval also produced a temporal trend less consistent with the lidar temporal trends in AOD compared to the h=0.6 km AODs (Fig. S14). Therefore, using an a-priori with h=1.2 km for the AOD retrieval appeared to produce a lower quality retrieval for Aug 23. See response to reviewer comment #2 for why an O_4 scaling factor was not applied.

8. Page 22, lines 1-3: You said that a major advantage of the MAX-DOAS technique is the simultaneous retrievals of total columns and vertical profiles of trace gases and aerosol extinctions without requiring a priori information. I am a bit puzzled by this sentence since a priori information is needed in the Optimal Estimation retrieval approach you used. A clarification is needed here.

Response: Agreed, this sentence was unclear. Sentence was modified Page 23 lines 1-3 to make it clear that the a priori information referred in this case to the aerosol characteristics that can be needed to retrieve accurate AODs from lidar compared to MAX-DOAS.

<u>Technical corrections: '->' denotes 'should be replaced by'</u>

1/'Honninger et al.' -> 'Hönninger et al.'

Response: Corrected throughout.

2/'Clemer et al.' -> 'Clémer et al.'

Response: Corrected throughout.

3/The first sentences of both Sections 2.1 and 2.2 are a bit redundant. I would start Section 2.1 by 'The MAX-DOAS instrument was operated at an elevation of ~10 m. . ..' and Section 2.2 by 'The MAX-DOAS instrument is a mini-DOAS spectrometer from Hoffmann Messtechnik GmbH measuring scattered sunlight. . ...' or something similar.

Response: Updated based on reviewer comments on page 5 on lines 3 and 18.

4/Page 5, line 25: 5◦C (C not in superscript)

Response: Corrected on page 5 line 26.

5/Page 22, line 1: 'total column' -> 'total columns'

Response: Corrected on page 23 line 6.

6/List of References: 'Atmospheric Meas. Tech.' -> 'Atmos. Meas. Tech.'; Same 'Atmospheric Chem. Phys.'

Response: This has been corrected throughout the list of references on pages 49 to 55.

7/Legends of Figures 4-9: You should add a short description of the error bars presented in the plots.

Response: A short description of the error bars was added to the last sentence in the captions of Figures 4-9.