Interactive comment on "The First Evaluation of Formaldehyde Column Observations by Pandora Spectrometers during the KORUS-AQ Field Study" *by* Elena Spinei et al.

Elena Spinei et al.

eslind@vt.edu

We thank the reviewer for his/her thoughtful and constructive comments/recommendations.

< The description of the ground-based measurement technique (Sections 2.2) is insufficient. The Hottle et al. (2009) reference is for the LIF technique, which was not used here. Please include a more relevant reference and a more detailed description. More information on the instrument accuracy should be included, similar to the discussion of the CAMS measurement (page 13, lines 5-8). >

- We have expended the in-situ surface measurement description and added more appropriate reference: " Surface HCHO concentrations were measured at Mt. Taehwa and Olympic Park by tunable infrared laser direct absorption spectroscopy (Li et al., 2013) at mid-IR wavelengths (QC-TILDAS from Aerodyne Research, Inc). In situ HCHO measurements were conducted by the U.S. Environmental Protection Agency (EPA) at the Olympic Park Research Site, and by the Aerodyne Research, Inc. at the Mt. Taehwa Site.

Light from 1765 cm-1 (Olympic Park) and 2831.6 cm-1 (Mt. Taehwa) quantum cascade lasers were passed through a 0.5 liter absorption cell with an effective path length of 76 m. Air was sampled at 12 liters per minute from a heated glass inertial inlet system located at a height of around 10 and 15 meters above ground level for Olympic Park and Mt. Taehwa sites, respectively. The inertial inlet kept particulate matter greater than 100 nm out of the absorption cell in the instrument. A critical orifice controlled the instrument flow rate. The pumping speed dictated the cell pressure (35-45 torr). All tubing between the inertial inlet and the measurement cell was Teflon and heated to 30C.

Absorption measurements were made relative to a zero air background gas obtained from an ultra-high purity zero air gas cylinder. Backgrounds were taken through the same inertial inlet that samples were measured. A 30 second background (with a 15 second flush time) was taken every 10 – 15 minutes. Nitrogen (N2) was flowed constantly through a permeation tube heated to 50C to provide a reference gas. This was added to the sample stream for 90 seconds every 15 minutes as a standard addition to monitor instrument stability over time.

Spectra were averaged for 1 second intervals and fit using a non-linear least squares fitting algorithm, with parameters based on the HITRAN database (CITE). 1-second HCHO data was averaged to 10-second and 60-second averages to improve precision. Allan deviation (estimate for precision) for 10-second HCHO data is 0.100 ppb and for 60-second data is 0.060 ppb. Estimated accuracy is approximately 10%."

Li, J. S., Chen, W. and Fischer, H.: Quantum Cascade Laser Spectrometry Techniques: A New Trend in Atmospheric Chemistry, Applied Spectroscopy Reviews, 48(7), 523–559, doi:10.1080/05704928.2012.757232, 2013.

<Specific comments:> <Page 2, line 20: Is the absolute bias greater than 16%? The slope is 1.16, but given the positive intercept, I would expect the bias to be greater. >

- We have reworded to make it clearer that Pandora overestimates the columns: "...showed that Pandora values were larger by 16% with a constant offset of 0.22 DU"

<Page 3, line 20: Add more recent references for global HCHO inversions (e.g. Bauwens et al., 2016). >

- Added

<Section 2: Please reference a figure which shows the two locations on a map. This could be figure 4 with a zoomed-out key.>

- We added Fig 1(b) with the locations and the estimated photon paths through the mixing layer.

Figure 1. (a) Schematic of direct sun observation geometry; (b) Estimated photon path through mixing layer derived from Vaisala Ceilometer CL51 backscatter profiles (910 nm) over Olympic Park and Mt. Taehwa during KORUS-AQ. Color coding represents time of the day (blue – morning (horizontal distance 1.5 km) and red - evening (horizontal distance 6 km)).

<Section 2.1: Please state the integration time.>

- The following was added: " Direct sun spectra are taken at variable integration times (2.5 ms to 4 s) with the total measurement duration of 40 s."

<Page 9, lines 6-11: Line 7 says fitting scenario can result in an error of +/- 10%, and line 10 says the SCDS varied by 20%. Please clarify/simplify.>

- The following sentence was deleted: ", but varied by 20% in HCHO △SCD. The
selected fitting scenario was in the middle of the spread."

<Page 9, line 19: Add "totaling to 14%" here or elsewhere in this paragraph. Also, is
the 9% error from the HCHO cross section itself should be noted somewhere in the
text (not just the table). >

- The following sentence was modified: "Pinardi et al. (2013) reported that for multi-axis
DOAS geometry (336.5-359 nm), error due to σ(O3) selection (Bogumil et al., 2003
vs. Malicet et al., 1995) can result in HCHO △SCD error of 13% (△SCD); σ(NO2)
selection (Vandaele et al., 1998 vs. Burrows et al., 1998)) - up to 5%, and σ(BrO)
selection (Fleischmann et al., 2004 vs. Wilmouth et al., 1999) - about 2%, totaling
14%. Uncertainty in HCHO cross section is 9%."

<Page 9, line 23: If possible, please state if these errors are expected to be large or
small in comparison with the other listed uncertainties in △SCD.>

- The following was added: "Figure 2(b) shows an example of common optical depth
residuals calculated by the DOAS fitting algorithm of 4537 cloud/spatial stray light free
DS measurements and scaled by DS AMF. Figure 2(c) illustrates the effect of this
residual spectrum on the retrieval of 0.5 DU (background levels) of HCHO. Some of
this common residual spectrum is potentially due to ESS. At this point we estimate that
the error due to ESS is on the order of 0.025 DU."

<Figure 4: Simplify x-axis notation (ppt x E3 to ppb)>

- The change was made.

Page 30, final conclusion: It is worth noting here that the Pandora instrument performed
about as well as an in-situ measurement combined with measured mixing layer heights
(Table 3).

- The reviewer's recommendation is addressed in conclusion No. 9: "Comparison
between Pandora and "ground-up" columns over Olympic Park suggested that profile

shape (4) with measured MLH and exponential decay produced the best agreement (slope = 1.03 $\pm$ 0.03, intercept = 0.29 $\pm$ 0.02 DU and R2 = 0.78$\pm$0.02). The source of the offset bias is not clear at this point. These results suggest that reasonable estimation of the surface concentration can be done from the total column HCHO and MLH data. "

<References: Bauwens, M., Stavrakou, T., Müller, J.-F., De Smedt, I., Van Roozen-dael, M., van der Werf, G. R., Wiedinmyer, C., Kaiser, J. W., Sindelarova, K., and Guenther, A.: Nine years of global hydrocarbon emissions based on source inversion of OMI formaldehyde observations, Atmos. Chem. Phys., 16, 10133-10158, https://doi.org/10.5194/acp-16-10133-2016, 2016.>
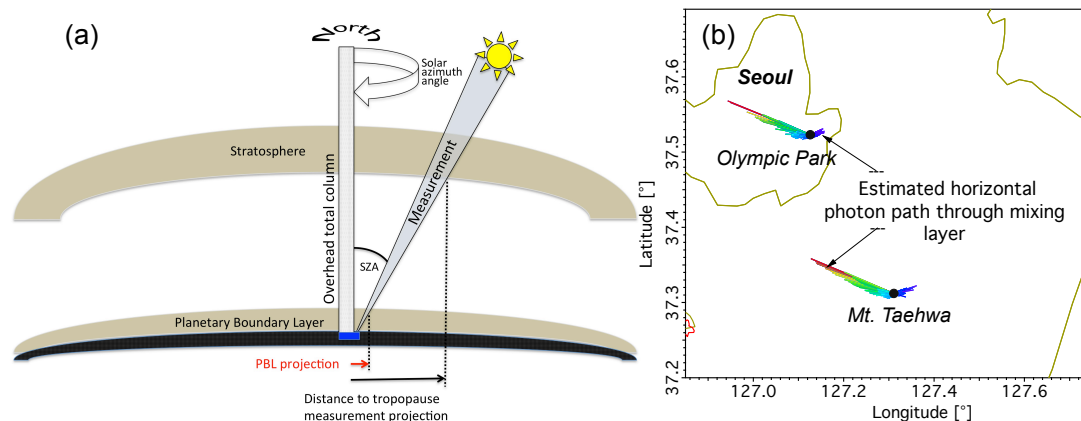
Fig. 1. Figure 1 (see text for caption)