Atmospheric
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Techniques

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Discussions

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

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<General comments

1. The introduction part emphasizes the importance of HCHO on atmospheric chemistry. However, I think it is also important to describe the current status of Pandora HCHO measurements (e.g., advantages, drawbacks, uncertainties, etc.), especially the necessity of performing inter-comparisons. >

- We have moved/added the following paragraphs to the introduction:

"The uncertainties of the DOAS derived HCHO columns are impacted by the DOAS fit

uncertainty and the uncertainty in the air mass factors. Validation of such measurements is challenging due to air volume sampling differences between different platforms. In this paper we present HCHO total columns from DOAS measurements of unscattered, direct-sun photons using NASA/GSFC (National Aeronautics and Space Administration / Goddard Space Flight Center) Pandora instruments and in-situ measurements over two sites during the Korea-United States Air Quality Study (KORUS-AQ) conducted in May-June 2016 in South Korea.

Pandora instruments are field grade spectroscopic UV-VIS systems (Herman et al. 2009). They are part of the growing joint NASA (USA) and European Space Agency sponsored Pandonia Global Network (PGN). The main goal of PGN is to provide consistent ground-based total NO2, HCHO and O3 columns for satellite validation. The major advantages of PNG include uniform instrument design, calibration, and centralized data monitoring, processing and distribution. Direct sun observation geometry eliminates the need for atmospheric radiative transfer modeling and simplifies data interpretation. PGN currently operates 75 instruments and is expected to have about 300 instruments by 2020-2021. Their product quality assurance is extremely important for satellite validation.

Pandoras deployed during KORUS-AQ were retrofitted with new UV grade fused silica windows with broadband antireflection coating (ARC, 250-700 nm). This modification from the earlier versions of Pandora (pre 2016) was necessary to decrease spurious spectral structure in direct sun (DS) spectra. This new ARC window improved NO2 and O3 measurements and made HCHO retrieval from Pandora DS measurements possible for the first time. KORUS-AQ Pandora measurements are extensively evaluated with both ground-based and airborne in situ observations of HCHO available during this study. "

<2. I suggest that the authors consider to reorganize the manuscript in the way that the methodologies, the results, and the discussions are put in individual sections. Within the current structure, I feel the readers could be messed up with informations

such as how to convert mixing ratios to VCDs, how the measurement uncertainties are estimated, but possibly be distracted from the major concern, i.e., what the inter-comparison looks like and the reason for disagreement. >

- We agree that there is a lot of information about calculation details in the paper. To help reader navigate we added a "paper road map" at the end of the introduction and renamed/numbered some of the sections:

"The rest of the paper is organized in the following sections. Section 2 describes in detail ground-based (Pandora and in situ) and aircraft measurements during KORUS-AQ 2016 study. Section 3 explains how HCHO vertical column densities are calculated from the in situ measurements (aircraft and surface) for comparison with Pandora column measurements. Section 4 shows the results by comparing HCHO vertical columns from Pandora, surface and aircraft measurements. Section 5 focuses on conclusions"

<Specific comments>

< Line 18 – 25, Page 7: The AMF is calculated from a geometric estimation. If considering the effect of aerosols and clouds, how large would the AMF change? >

The main advantage of direct sun measurements is to simplify AMF calculation and eliminate the need for aerosol and cloud radiative transfer modeling. Therefore, the goal is to exclude cloudy measurements by filtering the data based on the irradiance measurement error, residual fitting OD RMS and relative HCHO error. Since the Ring pseudo cross section was not fitted in the DOAS analysis any significant contribution of the scattered light to the direct beam would result in increase of the above parameters and the data would be excluded from this analysis. To confirm that the spatial stray light did not affect the AMF of the remaining data we also compared O_2O_2 vertical column densities (VCD) derived from the DOAS fit ($332 - 385$ nm) using geometrical AMF and VCD from the radiosonde measurements. They agreed within 7% on average (dSCD slope vs. $AMF_{geometrical}$ was 1.35×10^{43} molecules2 / cm5, $R^2 = 0.98$, see Fig.1, radiosonde average VCD was 1.28×10^{43} molecules2 /cm5). Point-by-point Pandora

measured O_2O_2 VCD vs radiosonde integrated O_2O_2 VCD can vary by +/- 13%. While there is a chance that some of this disagreement is due to AMF calculation, most of it is due to DOAS fitting challenges (cross section uncertainties, temperature dependence, etc)

<Line 17, Page 23: As the authors described, the day to day agreement between HCHO VCDs derived from the three techniques varies a lot. The authors explain this as a result of "spatial and temporal heterogeneity". I think the authors should spend more efforts on digging out the exact reason. >

- While we tried to understand the reasons for the differences we cannot do it using the data presented in this paper. Other studies are currently being conducted that look at chemical transport models and species distribution accounting for emissions and chemistry. Considering the complexity of the problem we think it is outside of the scope of this paper.

< Would it be possible for the authors giving some estimates on the heterogeneity of HCHO distribution? In Page 16, the authors has checked the spatial heterogeneity and only found a difference of less than 20%. >

This topic is partially covered in Section 2.3:

"Figure 3 shows time coincident in situ surface HCHO vmr at Olympic Park and Mt. Taehwa. The average vmr during the campaign at Mt Taehwa was 2.68±1.45 ppb (min=-0.74 ppb, max=9.22 ppb, median=2.39 ppb, Q25=1.59 ppb, Q75=3.51 ppb). Somewhat higher vmr were observed at Olympic Park: 3.46±1.59 ppb (min=0.07ppb, max=12.73ppb, median=3.35ppb, Q25=2.38ppb, Q75=4.40ppb)."

The time coincident ground-based in-situ measurements show that HCHO vmr difference can be on the order of 30-50% between the two sites. Meaningful discussion of the heterogeneity of HCHO distribution will require looking at more than two locations at the same time. Unfortunately, Mt. Taehwa and Olympic Park were the only sites with

HCHO monitors. More in-depth analysis of other chemical species in combination with chemical transport models will be required to address this question.

< Considering there were not time lag between the three techniques, I could not understand how temporal heterogeneity come from. >

Aircraft measurements took on the order of 30 min to cover the area within 15-km radius and spiral to 8 km altitude, so the measurements are not truly time coincident. Since HCHO is photo chemically produced in the atmosphere and depends on the presence of anthropogenic emissions, and also is impacted by availability of solar radiation for reaction and mixing, temporal heterogeneity is expected. DC-8 measurements showed that HCHO vmr profiles over the two sites were different depending on the time of day. Ceilometer backscatter data also showed complex PBL evolution in early mornings witch is expected to impact HCHO distribution within short time intervals.

<Could it be the uncertainty of the Pandora measurements was underestimated under some circumstances? >

- The DOAS fitting residual OD rms are similar in the final filtered data. One of the potential causes that have not been considered in this paper is change in atmospheric effective temperatures of different species (e.g. O_3, HCHO, O_2O_2) on the DOAS fitting of HCHO. This will be focus of the future work

<And could it be the AMF for the days with large discrepancy been under-/over estimated?>

- The data were strictly filtered and remaining error in AMF cannot account for such large differences

<Line 23–24, Page 25: It is unclear to readers how the number -11% and -19% is calculated. >

- The following sentence was added: "Heterogeneity and altitude "undersampling" errors were added in quadrature, assuming their independence"

C5

< Why DC-8 measurements could underestimate HCHO mixing ratio? The comparison between the ground-based and the airborne in-situ measurements were not performed exactly side-by-side. Therefore, in my opinion, the slope of the linear regression in Fig.5 does not really indicate that DC-8 measures lower values as described in Line 16.>

- We agree and do not claim that DC-8 underestimates the vmr measurement itself but rather that there is spatial heterogeneity in HCHO distribution within a 200-470 m volume that causes this difference between DC-8 and ground-based measurements.
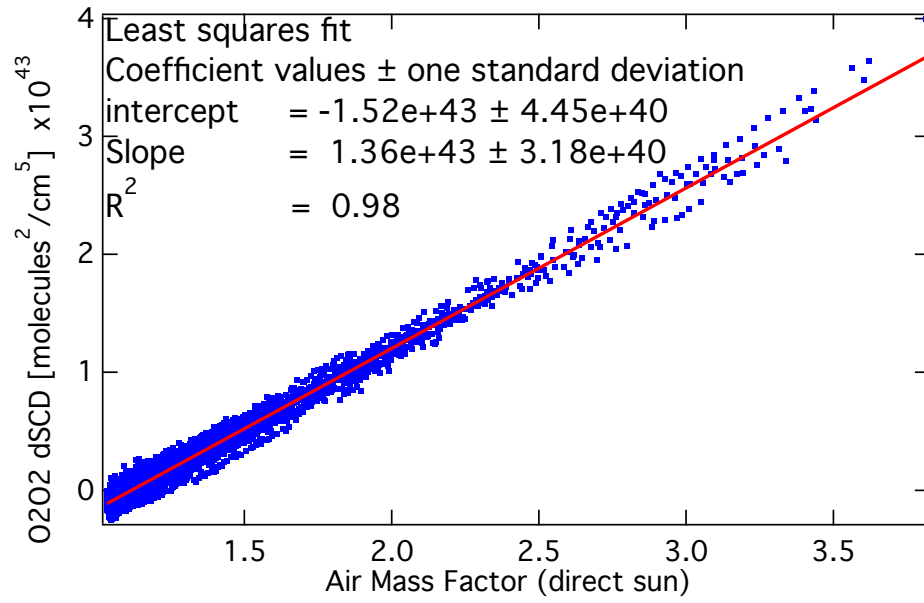
<Section 6, Page 28: I suggest the authors to re-write this part. (1) Separate the discussion and the conclusion. (2) Give concise conclusion by providing major findings.>

We feel that the conclusion-discussion section is focused right now.

Fig. 1. Langley Plot of dSCD(O2O2) retrieved in 332-385 nm wavelength window vs. direct sun AMF (geometrical).