

Interactive comment on “NO₂ and HCHO measurements in Korea from 2012 to 2016 from Pandora Spectrometer Instruments compared with OMI retrievals and with aircraft measurements during the KORUS-AQ campaign” by Jay Herman et al.

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Please note the color coding. Green was for Referee 1 and Gray for referee 2. I used bold for replies to referee 2 that are not inserted into the paper.

A formatted version is in the Supplement

Interactive comment on “NO₂ and HCHO measurements in Korea from 2012 to 2016

C1

from Pandora Spectrometer Instruments compared with OMI retrievals and with aircraft measurements during the KORUS-AQ campaign” by Jay Herman et al. Anonymous Referee #2 Received and published: 10 June 2018

Specific comments: Abstract, line 44: What kind of average is meant by the mentioned “PSI C(NO₂) averages” and “OMI averages”? PSI C(NO₂) 30-day running averages Introduction: Please mention that these are direct sun measurements. I added: “The intent of the network was to integrate direct-sun column density observations of NO₂ and HCHO into a multi-perspective framework of observations including ground-based, satellite, and airborne measurements of air quality.”

The objective and aim of the campaign should be declared more clearly in the text. Done Although the cited reference Spinei et al., 2018 discusses the analysis procedure very extensively, please mention in the introduction at least briefly the retrieval algorithm (DOAS?), what kind of reference spectrum is used and how the measured slant columns from the direct sun measurements are converted into vertical columns. The following has been added “The retrieval algorithm is based a direct-sun spectral fitting method similar to the well-accepted DOAS (Differential Optical Absorption Spectroscopy, Platt, et al., 1979 and Platt, 1994). NO₂ absorption cross sections were obtained from the laboratory measurements of Vandaele et al., 1998, and HCHO cross sections from Meller and Moortgat (2000). The PSI reference solar spectrum is constructed from a high resolution extraterrestrial spectrum from 270 nm to 1000 nm merged from different sources (Bernhard et al. (2004). Solar spectrum sources are from: Kurucz (2005) normalized to Thuillier et al. (2004), SUSIM/Atlas-3 spectrum (VanHoosier et al., 1996), and the spectrum from Gueymard (2004). One of the advantages of using direct-sun observations is the accurate conversion to vertical column based on a geometric calculation of the slant path air mass factor AMF for a known solar zenith angle SZA is with a slight correction to the function Secant(SZA) (Herman et al., 2009 eqn. 3). A complete description of the retrieval algorithms and PSI operations are given in the PSI software manual (Cede, 2017). Accuracy in the DOAS-type

C2

retrieval is obtained using careful measurements of the spectrometer's slit function, wavelength calibration, knowledge of atmospheric absorption cross sections, and the solar spectrum at the top of the atmosphere. Accuracy for C(NO₂) has been estimated to be ± 0.05 DU. A recent addition of anti-reflection coatings to the PSI optics has improved accuracy and precision by reducing the residuals associated with spectral fitting using trace gas absorption cross sections. The reduced residuals are necessary for the retrieval of formaldehyde and bromine oxide that absorb in spectral regions dominated by ozone and NO₂. Other DOAS-type measurements have been made in Korea based on observations of sky radiance ratios (e.g., Multi Axis MAX-DOAS: Kanaya, et al., 2014) and direct-sun DOAS using a PSI in Seoul, Korea (Park et al., 2018)."

Since it is mentioned later on in Section 2 for the measurements in Anmyeondo, please discuss briefly the contribution of the stratosphere to the presented total vertical columns. How large is the contribution and does it change over the day? Can stratosphere and troposphere be separated? The measurement from the PSI is for total column (stratosphere + troposphere). For NO₂, the stratospheric contribution is approximately 0.1 DU and it does change during the day by about 0.05 DU. The direct sun measurement does not permit separation of troposphere and stratosphere. A sentence was added, "Of these sites, Anmyeondo frequently (40%) retrieves values of C(NO₂) that are close to the typical stratospheric values of 0.1 ± 0.05 DU. Other sites occasionally have clean days with similar low values."

Please mention briefly what other work on ground-based (DOAS type) Korean/Asian air pollution measurements has been published in other studies in the past to put the aim of the campaign/study into context. There are other DOAS measurements, but they are all MAX-DOAS, which are quite different than direct sun. I added a brief comment, "Other DOAS-type measurements have been made in Korea based on observations of sky radiance ratios (e.g., Multi Axis MAX-DOAS: Kanaya, et al., 2014) and direct-sun DOAS using a PSI in Seoul, Korea (Park et al., 2018)."

Fig. 2a, page 5: The agreement between the instruments is quite impressive. But

C3

what happened at 17:30-18:00 local time? Are these real differences or has the Pan27 instrument some missing data gaps and the connected data points convey a wrong impression? These are differences caused by low signal from increasing cloud cover that affects the retrieval algorithm. There are small differences between instruments that introduce different amounts of noise in the signal for small signals. Section 2, line 195f, page 8: See comment on stratospheric contribution above. The most frequently occurring C(NO₂) value at Anmyeondo is 0.15 – 0.2 DU, which means that the measured NO₂ amount are partly from the stratosphere (0.1 ± 0.05 DU) with very little tropospheric or boundary layer NO₂. There are occasional C(NO₂) plumes that could be from industrial activity to the north, and, perhaps, from China. Transport of NO₂ from China occurs episodically in significant amounts (Lee et al., 2014).

Section 3: Please explain in more detail why the observed NO₂ daily patterns fit so well to automobile and power generation emissions. The pattern measured by PSI in Korea during the spring KORUS-AQ campaign differed from other non-Korean locations in that there usually a weaker morning peak in Korea compared to the afternoon. The strong afternoon peak does not occur every day, but is often enough to be notable. The meteorological effects certainly play a role, but at this time the appropriate model studies are not available.

In many of the studies on the NO₂ diurnal cycle in polluted urban regions two NO₂ peaks in the morning and afternoon are observed corresponding to the morning and afternoon traffic rush hour. Do you have an explanation why the morning rush hour is hardly visible in the presented measurements and why the afternoon peak is so pronounced? Do you also observe a weekly cycle in your NO₂ measurements, like it has been observed in polluted regions and discussed for example in Beirle et al. (GRL, 2003) or Ialongo et al. (AMT, 2016)? Yes, there is a weekly cycle in that Sundays usually have less pollution than Wednesdays. Section 4: About the seasonal cycles: Please discuss briefly why the seasonal cycle of NO₂ has its minimum in August/September and its maximum in winter/early spring. Is there more heating during

C4

winter times in Korea or is it just due to less OH radicals because of less light in winter? Winter in Korea is complicated with quite a bit of cloud cover and precipitation. This certainly would affect the chemistry. Plus the cold winter weather certainly increases the amount of energy used, which produces more NO₂. Since I did not do the model studies, I preferred to just leave the data without a “hand waving” explanation. Section 4, line 291, page 13: The “strong effect on local air quality” is an improvement of local air quality, right? The strong effect on local air quality mentioned in the paragraph concerns large amounts of total column NO₂, most of which is near the surface. This probably makes the air quality worse. Section 6, line 531f, page 27: What is the reason for this seasonal increase during May and June? Unfortunately, there are no long term measurements of C(HCHO), so there is no way of determining if the increase is from sources or VOC chemistry. The PSI was not capable of measuring HCHO prior to late 2016. Technical corrections: Line 43, page 2: please add “local time” or LT Added: “OMI overpass local times (LT = 13.5 ± 0.5 hours).” Line 48, page 2: please enclose “FOV” in brackets OK Tab. 1: The degree symbol is missing for latitude and longitude values OK Line 229, page 10: Please round off the values for H₂O and CO₂. Six significant(?) digits are unnecessary here, since this paragraph is only about getting a general impression on the order of magnitude of the emissions from automobiles. OK “containing H₂O (144 ppm) and CO₂ (122 ppm)”. Fig. 6: Between Panel A and Panel B the x axis tick labels A, M, J and J (April, May, June, July) are missing. Line 298, page 13: please add “local time” or LT Fig. 9a: Seoul (left panel): Why does the 3-month average (solid lines) show values where around 6 month of data are missing? Or is it just a linear interpolation between the values before and after the gap? Missing data are represented by linear interpolation in the plots, but not any analysis. I have added a statement in the caption. “Fig. 9a Comparisons between the daily values of C(NO₂) for OMI (black) and PSI (red) at Seoul and Busan for a 5-year period. Solid lines show the average seasonal variation (Lowess(0.1)), see also Fig. 9b. Linear interpolation is used where there are missing data points.” Line 429, page 19 and Fig. 14: The numbers in “Integ(0.026, 7.2)” are given in kilometers? Yes. Added: “Integ(0.026, 7.2

C5

km)”

Tab. 2.: “PSI HCHO” is missing the “DU” (like in “DC8 HCHO DU”) Table 2 is now Table 2 Taehwa Mtn DC8 compared to PSI measurements (see 10 Jun in Fig. 18) Date LT DC8 HCHO DU PSI HCHO (DU) Percent 11 May 08:25:19 0.4 0.6 67 18 May 08:34:26 0.4 0.5 80 30 May 12:05:00 0.5 0.9 56 10 Jun 08:22:45 1 1.16 86 10 Jun 12:22:53 1 1.5 67 10 Jun 15:46:03 1 1.3 77

Line 545, page 29: “... very high amounts of urban pollution from NO₂ and HCHO *, and more moderate, but still high values, away from the urban centers.” *close to the urban centers

Changed to “, but still high values in Mt Taewha and Yeogju, which are some distance from the major

Please also note the supplement to this comment:

<https://www.atmos-meas-tech-discuss.net/amt-2018-56/amt-2018-56-AC2-supplement.pdf>

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2018-56, 2018.

C6