Atmos. Chem. Phys. Discuss., 14, C9196–C9200, 2014
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Atmospheric Chemistry and Physics Discussions

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14, C9196–C9200, 2014

Interactive Comment

Interactive comment on "Meteorological controls on the vertical distribution of bromine monoxide in the lower troposphere" *by* P. K. Peterson et al.

Anonymous Referee #1

Received and published: 15 November 2014

"Meteorological controls on the vertical distribution of bromine monoxide in the lower troposphere" by P. Peterson et al. describes an optimal estimation approach to retrieve only 2 pieces of information (vertical columns within 200m and 2km) of BrO instead of the traditional full profile retrieval and evaluates effects of surface temperature, atmospheric stability, wind speed and aerosol loading on the BrO activation. This is a very important topic and is well suited within the scope of ACP. I recommend publishing the article after some modifications.

Main concerns. 1) MAX-DOAS analysis shown in this study is a two-step approach, where an "ad-hoc" optimal estimation (EO) is used to first retrieve the aerosol extinction coefficient profile and second - BrO profile. The 2nd step depends on the 1st step. Since the authors rightfully criticize applicability of the OE to the 2nd step where DOF is

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approximately 2 and full profile cannot be reliably retrieved, the same argument applies to the 1st step (same MAX-DOAS data are used). The authors have to demonstrate that the OE in the first step provide reliable enough profiles for the 2nd step and that the error from the aerosol extinction profile retrieval does not introduce a significant error in LT-VCD BrO analysis. In addition, the retrieved aerosol extinction profile is MAX-DOAS averaging kernel smoothed profile with low sensitivity above 1 km. The forward RT modeling in the second step requires the "true" profile. It will be interesting to see what effect this resolution degradation has on the LT-VCD BrO analysis. 2) Since BrO activation is linked to heterogeneous reactions any dependence on other physical parameters (e.g. temperature) should be done for the same aerosol conditions. Under strong wind conditions it is also important to know the source of air masses brought into the MAX-DOAS instrument field of view. Are there any independent aerosol measurements at the NOAA and/or ARM Barrow facilities that can give information about the dependence of wind speed and aerosols? In general, Arctic climatology is very complicated and decoupling of different meteorological parameters is not straightforward. Please also include information whether there were passing polar lows and/or cyclones.

Detailed comments:

p. 23954 , l 24. The sensitivity to upper troposphere and lower stratosphere is reduced but still present especially at SZA > 75deg. How do the authors account for it in the analysis at SZA > 75deg?

p. 23954 Methods, MAX-DOAS measurements: I suggest that all the data presented in this work (IL1, BARC, SIMS, sondes) are described in this section, including description of the measurement locations and azimuthal direction of MAX-DOAS measurements. It will also be helpful to get an idea about the meteorological conditions during the campaign and description of the air masses that are traversed by the measured photons. How do you treat cloudy data?

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p. 23955 l. 4 How well is the optical bench temperature maintained? Ocean Optics spectrometers tend to have strong temperature dependence of the slit function (resolution).

p. 23955, l. 15. Add “transfer” after instrumental

p. 23955, l. 20 It will be more meaningful to present residual OD RMS as a function of viewing elevation angle. Since DOAS fitting errors are linearly related to the residual OD RMS I suggest including BrO/O4 DOAS fitting errors as a function of elevation angle too.

p. 23956, l. 13. What is the a priori covariance matrix? What are the assumptions about the aerosol optical properties? Are you applying any correction factors to the measured dSCD(O4)? Could you estimate BrO 200m-VCD and LT-VCD errors due to aerosol profile error?

p. 23957, l. 25 How do you define “the layer in which the observer resides” from a MAX-DOAS perspective?

p. 23959, l. 19, Payne et al (2009) does not “describe how to calculate the BrO averaging kernel for the coarsened grid” the paper applies the method to methane not BrO. Please check your reference or rephrase the sentence. The cutoff 0.7 was used for satellite nadir measurements of methane. While it is likely it is applicable to MAX-DOAS ground-based measurements I would suggest doing some sensitivity studies to confirm this.

Payne, V. H., Clough, S. A., Shephard, M. W., Nassar, R., and Logan, J. A.: Information centered representation of retrievals with limited degrees of freedom for signal: application to methane from the tropospheric emission spectrometer, J. Geophys. Res., 114, D10307, doi:10.1029/2008JD010155, 2009. 23953, 23959

p. 23962, l.15 Please replace “observes” for “retrieves”. Technically O3 absorption is present in MAX-DOAS data.

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p. 23963, l. 25 What do you mean by "large changes in the environment in MAX-DOAS field of view"? Are there any significant differences in elevation, snow cover, landscape, and aerosol types? If there is a significant difference along the line of sight how did you treat it in RT modeling? Please show MAX-DOAS azimuthal direction on Figure 5.

p. 23964, l. 4 Figure 6 gives an impression that a significant number of time coincident observations exist. After closer inspection CIMS data are mainly shown for nighttime when MAX-DOAS instrument did not measure. Why there are so many "holes" in CIMS data?

p. 23964, l. 6 Could you please elaborate why do you claim that the differences in observations at IL1 and BARC explain differences between BARC and CIMS locations. What is the azimuth angle of BARC and IL1 MAX-DOAS observations? Where they pointing towards each other? What were the meteorological conditions at the 3 sites?

p. 23965, l. 10 You compare daily averaged MAX-DOAS data with the 15:30 AKSD sonde data. Were the meteorological conditions constant during any particular day? Would the conclusion change if comparison done using time coincident or +/- 1 hr data?

p. 23965, l. 19. Could you please explain what you mean by “The short diurnal time scales of the temperature gradient..”

p. 23965, Since halogen activation is related to heterogeneous reactions it might give more insight if any dependence on temperature is derived from the data bins collected under the same aerosol loading conditions. Since LT-VCD accuracy has a stronger dependence on the aerosol loading than 200m VCD I suggest also including data where only 200m VCD is shown.

p. 23967, l. 29. What is the typical PBL height during the measurement period at this location?

I am not sure what the authors want to convey by “Therefore, the diurnal pattern of

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the vertical profile of BrO appears to reflect boundary layer dynamics. The observation of Pöhler et al. (2010) of decreasing surface BrO mixing ratio at higher temperatures could therefore reflect increased vertical mixing of surface sourced BrO that dilutes the surface concentration upon warming rather than being indicative of decreased halogen activation chemistry at higher temperatures."

If the source of BrO is located at the ground and shallow PBL is present (< 1000m, probably 400-600 m) convective mixing to the upper layers should not result in increase in measured LT-VCD (0 – 2000m). This implies that there is another source of BrO at altitudes > 200 m that is coincident with the potential PBL diurnal evolution. Do you have an independent PBL height estimation to confirm your statement about PBL diurnal evolution? It might be helpful to separate data into "colder" and "warmer" periods to see if the trend persists.

Role of wind speed (4.2) and Relationship between activation and aerosol particles (4.3) might not be reliably established from LT-VCD or VCD200m/LT-VCD since LT-VCD retrieval is potentially effected by the aerosol retrieval in the 1st step and due to decreased sensitivity of MAX-DOAS at higher AOT.

Figure 5. Please indicate MAX-DOAS azimuthal orientation. In addition, information about snow/ice cover will be valuable

Figure 7. Please show error bars for both MAX-DOAS and CIMS data

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