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Continuous observations of atmospheric HCHO by MAX-DOAS at Yokosuka, Japan: Verification and correlation with ozone

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Tropospheric ozone is an important greenhouse gas with the third largest global warming effect after CO2 and CH4. Its photochemical production in the atmosphere is not quantitatively understood, partly because of the large uncertainties in the amount and origins of the volatile organic compounds (VOCs) in the atmosphere serving as precursors of ozone. Accurate observations of formaldehyde (HCHO), formed from VOC oxidation simultaneously with ozone, would provide pivotal information of the VOC emission rates and ozone production mechanisms. This study focuses on the measurements of HCHO in the urban area, specifically at Yokosuka (35.32 degN, 139.65 degE), Japan. Appropriateness of the observations was verified and then the correlation with ozone concentrations was studied.

Differential slant column densities of HCHO were derived from MAX-DOAS spectrum observations (336.5 - 359 nm) at Yokosuka and then converted to the vertical column densities/profiles using aerosol information retrieved simultaneously. We obtained observational data from October 2007 to December 2013, and from 08H (0800-0900 JST) to 15H in winter and from 06H to 17H in summer. The HCHO concentrations at the lowest layer derived from MAX-DOAS agreed quite well with the monthly ground-based observations of HCHO conducted at Oppama site, 2 km west of our location. Although we showed that recent satellite observations of HCHO provided reasonable agreement with MAX-DOAS over a rural site near Moscow, Russia, poorer agreement was obtained at Yokosuka, potentially affected by the spatial inhomogeneity in the HCHO concentrations in or near the urban region.

Monthly averages of the tropospheric vertical column density of HCHO derived from MAX-DOAS at 13H showed clear seasonal variation with maxima during July-September ((1.4-2.0)x10ˆ16 molecules cmˆ-2) and minima during December-March. The temporal variation during summer was quite similar to that of ozone concentrations observed at Oppama, while the HCHO levels were quite low in April/May, when the ozone concentration was even higher. The HCHO partial column in the lowest 1 km altitude range at 13H during June-August showed tight positive correlation with the surface ozone concentrations. These analyses suggested that the MAX-DOAS observations of HCHO are successful in quantifying HCHO secondarily produced in the atmosphere by photochemistry and that the short-lived HCHO would be a useful tracer to differentiate in-situ ozone production from long range transport.

Keywords: Tropospheric photochemistry, Volatile organic compounds, Urban atmosphere, Ozone precursors