

Elucidating the Causes of Variations in Sulfur Dioxide Concentrations in the Planetary Boundary Layer in Japan Using the MAX-DOAS technique

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Climate change has become a global issue in recent years, with projections of 40 million deaths by the end of this century (United Nations Development Program). The greatest uncertainty in the effective radiative forcing is caused by aerosols, typically sulfates, produced from sulfur dioxide (SO₂) in the atmosphere (IPCC AR6). SO₂ not only contributes to climate change, but also causes air pollution and health hazards. Although SO₂ has been mainly observed at the surface, there is a lack of long-term observation in the planetary boundary layer (PBL), where a marked increase in concentrations having more spatially representativity occurs, and a systematic understanding of the causes of PBL SO₂ concentration fluctuations has not been achieved. In this study, we conducted long-term continuous observations using Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS), which is one of the ground-based remote sensing methods. The five observation sites are Sendai, Tsukuba, Chiba, Kasuga, and Fukue, which have different geographical features in Japan. The observation period was different at each site, with the Kasuga site being the longest continuous observation period of 9 years from 2014 to 2022. Measured spectra in a UV region of 310-320 nm were analyzed using the DOAS method to derive average SO₂ concentrations in the 0-1 km altitude layer with a horizontal scale of about 10 km. For Kasuga and Fukue, back trajectory analyses strongly suggested the influence of volcanic eruptions that brought daily SO₂ concentrations 10 times higher than the medians over the entire periods of observations. At the Kasuga site, the MAX-DOAS Δ SCD (difference in slant column concentrations between low and reference elevation angles) and LiDAR data supported the results of the back-trajectory analysis. On the other hand, the high concentration days at Chiba, Tsukuba, and Sendai sites strongly suggested the influence of power plants and factories around ports and coastal areas. At the Chiba site, four MAX-DOAS instruments were observed pointing in four different directions (north, east, west, and south). Of these directions, the south direction showed the highest values, but we also found a sharp decrease after September 2019. It was inferred that this factor was related to the shutdown of the Anegasaki thermal power plant located in the south direction of the Chiba site due to its aging and the reduced operation of the steel mill in Kimitsu due to the reduced demand for iron in the COVID-19 disaster. At these sites where anthropogenic influences were suggested, SO₂ concentrations were less than 10 times the median value during the observation period, even on the day when SO₂ concentrations were highest. This indicates that volcanic eruptions have a greater impact on the daily mean SO₂ concentrations than anthropogenic sources.

Keywords: MAX-DOAS, remote sensing, SO₂, air pollution, planetary boundary layer