Atmospheric Research 168 (2016) 80–91

Contents lists available at ScienceDirect

Atmospheric Research

journal homepage: www.elsevier.com/locate/atmos

Characteristics of formation and growth of atmospheric nanoparticles observed at four regional background sites in Korea

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ARTICLE INFO

Article history:
Received 21 January 2015
Received in revised form 25 August 2015
Accepted 25 August 2015
Available online 12 September 2015

Keywords:
New particle formation
Asia continental outflow
SMPS
Regional scale
CSEOF

ABSTRACT

Measurements of the number concentration and size distribution of atmospheric nanoparticles were conducted at four sites on the west coast of the Korean Peninsula by using identical scanning mobility particle sizers (SMPSs) in October 2012. The new particle formation and subsequent growth (NPF) of atmospheric nanoparticles, which were identified by the cyclostationary empirical orthogonal function (CSEOF) analysis technique, was observed on 11 out of 21 days at the Baengnyeong-do Comprehensive Monitoring Observatory (BCMO); and on 10 out of 21 days at the Korea Global Atmosphere Watch Center (KGAWC) from October 9 to 29, 2012. We also observed NPF events for 9 out of 21 days at both the Gosan Climate Observatory (GCO) and the Jeju Comprehensive Monitoring Observatory (JCMO). During the study period, NPF was simultaneously observed for five days at all four sites, which indicates that the NPF event had a spatial extent of at least 540 km.

A cold, dry and cloud-free continental air mass originated from northern China, formed favorable environmental conditions (e.g., increasing solar insolation at the surface) on simultaneous NPF at the four sites. These synoptic weather patterns were closely associated with an extraordinary typhoon passing over the south of Japan. The mean values of particle formation rates at BCMO (1.26 $cm^{-3}\ s^{-1}$) and KGAWC (1.49 $cm^{-3}\ s^{-1}$) were relatively higher than those at GCO (0.39 $cm^{-3}\ s^{-1}$) and JCMO (0.74 $cm^{-3}\ s^{-1}$), however, the growth rate showed a similar level among four sites. An increase in the spatial homogeneity and inter-site correlation of atmospheric particles among the four sites was apparent for small particles (diameter < 30 nm) on simultaneous NPF event days.

1. Introduction

The nucleation and subsequent growth of atmospheric nanoparticles are important processes of accumulation-mode aerosol formation in the troposphere (Deshpande et al., 2014; Merikanto et al., 2009; Yu et al., 2008). These newly formed particles can grow into larger-sized particles, which can directly alter incoming solar radiation and act as potential cloud condensation nuclei (Kerminen et al., 2012; Kuwata et al., 2008).

The formation and subsequent growth of atmospheric particles have been observed under various environmental conditions and spatial scales (Arizabalo et al., 2015; Kanawade et al., 2014; Kulmala and Kerminen, 2008; Kulmala et al., 2004; Mbengue et al., 2014; Minguillón et al., 2015; Siingh et al., 2013; Wang et al., 2014; Zhao et al., 2015). Hussein et al. (2009) investigated long-term measurements of particle number size distributions at five widely separated background sites in Finland, Sweden, and Estonia and reported that new particle formation and subsequent growth (hereafter, NPF) events took place over the entire study region, where the combined horizontal distance among the sites was up to ~1500 km. Air masses transported from the Arctic region formed favorable environmental conditions for NPF (Nilsson et al., 2001a,b; Sogacheva et al., 2008). Jeong et al. (2010) determined that distinctive NPF events were apparent simultaneously at five rural and urban sites, which were up to 350 km away, in Ontario, Canada. These regional NPF events were closely associated with intense solar irradiance and less polluted, cold, and dry air masses. Simultaneous NPF events were also observed in the polluted urban and regional background stations, which were separated by approximately 120 km, in the North China Plain (Wang et al., 2013). The conditions of the NPF event in the North China Plain could be characterized as higher precursor vapor concentration and pre-existing particles, even in the regional background environment.

Many observation-based studies reported the formation and subsequent growth of particles in the atmosphere, both in polluted urban environments and in Asian continental outflow, where abundant precursor gases and pre-existing aerosols are prevalent (Kim et al., 2009, 2013, 2014; Lee et al., 2008; Song et al., 2010; Wang et al., 2014; Wu

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http://dx.doi.org/10.1016/j.atmosres.2015.08.020
0169-8095/

et al., 2007; Yum et al., 2007). Yum et al. (2007) reported that distinct NPF events occurred under cloud-free weather conditions induced by anticyclonic circulations positioned over northeast Asia. Song et al. (2010) reported that NPF events took place every five to six days in spring because of the periodic passage of a synoptic high pressure system. Lee et al. (2008) mentioned that most of the nucleation events were observed at the conditions of high ultraviolet-B (UV-B) irradiance and dry atmospheric conditions, or low relative humidity. Although these previous studies were concluded from single site observation, a very high probability of NPF occurring simultaneously in the Asian continental outflow region, including the Yellow Sea and the Korean Peninsula, was suggested based on the investigation of synoptic-scale meteorological conditions (e.g., Kim et al., 2009, 2013).

Simultaneous NPF events at multiple sites in the Asian continental outflow region and associated meteorological patterns have not been well investigated. This study focuses on investigating NPF events observed at four regional background stations in Korea in October 2012 and their characteristics, such as formation rate (FR), growth rate (GR), and condensation sink (CS). We elucidate the regional meteorological patterns on NPF events, including simultaneous NPF days observed at four stations, and non-NPF event days. An inter-site homogeneity in terms of the occurrence of NPF events and consistency in the temporal variability according to particle-size distribution is also presented.

2. Methodology

Aerosol number and size distributions were simultaneously measured by using a scanning mobility particle sizer (SMPS) at four regional background stations in the western coastal region of the Korean Peninsula during October 2012 (Fig. 1). Four sites are located in the major transport paths of aerosols and gas-phase precursors emitted from the Asian continent (Kim et al., 2007; Yoon et al., 2010). Fig. 1 shows the vertical column amount of boundary layer SO_2, averaged for October 2012, over East Asia obtained from the ozone monitoring instrument (OMI) on board the satellite Aura. The European Center for Medium-Range Weather Forecasts (ECMWF) ERA-Interim (Dee et al., 2011) geopotential height (m) and wind vectors (m s^{-1}) at a pressure level of 850 hPa were superimposed. The Baengnyeong-do Comprehensive Monitoring Observatory (BCMO; 37.97° N, 124.63° E, 120 m above mean sea level), which is located in the northwestern region of South Korea, is an ideal location to monitor polluted or clean air masses from North China and North Korea. The Korea Global Atmosphere Watch Center (KGAWC; 36.54° N, 126.33° E, 46 m above mean sea level), which directly faces the Yellow Sea, can be frequently affected by the air masses transported from North China. The distance between KGAWC and BCMO is approximately 220 km. Both the Gosan Climate Observatory (GCO; 33.29° N, 126.16° E, 72 m above mean sea level) and Jeju Comprehensive Monitoring Observatory (JCMO; 33.35° N, 126.39° E, 558 m above mean sea level) located on Jeju Island are considered ideal locations to monitor the regional background atmosphere in the East Asian region because there are no local industrial emission sources. GCO is located at the western tip of the island, whereas JCMO is located on the middle of Mount Halla. The distance between the two sites is approximately 20 km, and that between GCO/JCMO and KGAWC (BCMO, in parenthesis) is approximately 360 km (540 km). The four sites were established and maintained by Korea National Institute of Environmental Research (BCMO and JCMO), Korea Meteorological Administration (KGAWC), and Seoul National University (GCO).

At four observatories, identical SMPSs (manufactured by TSI, and model 3034) were operated with aerosol flow rates of 1.0 L min^{-1} and sheath airflow rates of 4.0 L min^{-1}. The size range was set to be 10–487 nm in mobility diameter (D_p; 54 bins) (Kim et al., 2013). In addition, sulfur dioxide (SO_2) concentration was measured at BCMO and

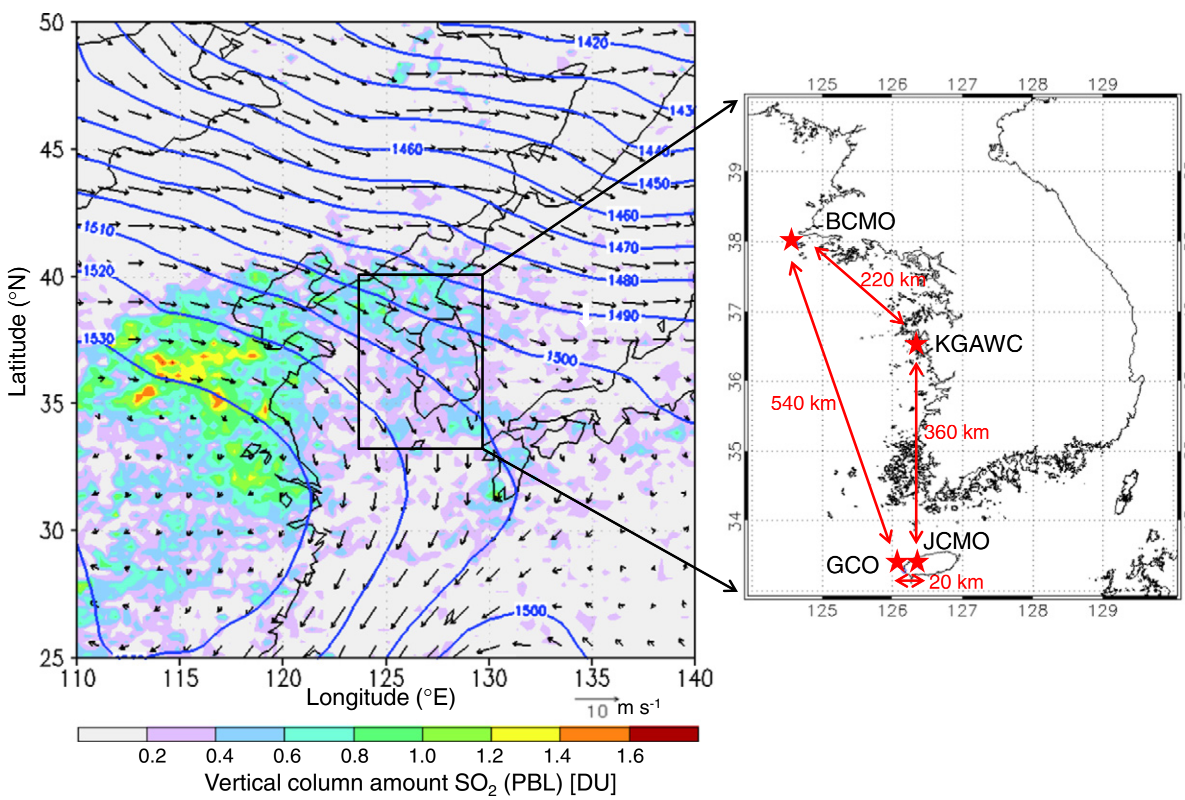

Fig. 1. (Left) Vertical column amount boundary layer SO_2 from the ozone monitoring instrument (OMI) averaged for October 2012 over northeast Asia. The European Center for Medium-Range Weather Forecasts (ECMWF)-Interim geopotential height (m) and wind vectors (m s^{-1}) at a pressure level of 850 hPa are superimposed. (Right) Enlarged map for the locations of the four observation sites (stars) in Korea; the distances between the sites are also shown.