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A numerical study on indicators of long-range transport potential for anthropogenic particulate matters over northeast Asia

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ABSTRACT

Several indicators of the long-range transport potential of particulate matters have been proposed. These indicators can be used in the interpretation of the long-range transport process over northeast Asia. In order to characterize the indicators of long-range transport potential for days during which the concentration of particulate matter is high, we first classified these days during which PM_{10} concentration was high into two extreme synoptic cases: long-range transport (LRT) case, and local emission with stagnation (LES) case. The classification employed here was on the basis of semi-empirical approach by employing the synoptic meteorological variables including relative vorticity, vorticity advection, and geostrophic wind speed/direction at a geopotential level of over 850 hPa. Simulation results of the two cases using the MM5-CMAQ modeling system were contrasted to determine suitable indicators of LRT potential.

The results showed that the most effective indicator is the ratio of aromatic organic compounds to NO_x, with correlation coefficients of 0.70 for toluene/NO_x, and 0.72 for xylene/NO_x. The ratios of N-containing species such as NO_x (or NO_y) to CO were the next best alternative indicators, with correlation coefficients of 0.53–0.62. The sulfur conversion ratios such as $SO_4^{2-}/(SO_2 + SO_4^{2-})$ and SO_4^{2-}/SO_2 suggested good indicators for high sulfate condition (i.e., $SO_4^{2-} > 10\ \mu g\ m^{-3}$). However, carbon aerosol ratio OC/EC did not show clearly separated regression lines distinguishing the LRT and LES cases, implying that ratios of both sulfur conversion in less polluted atmosphere and carbon aerosol should be considered carefully using them with confidence about their indication of the LRT potential over northeast Asia.

1. Introduction

Pollutants can be transported from one country to another depending on the prevailing weather conditions, which change daily. Identifying the origin of air pollutants, i.e., distinguishing long-range transport (LRT) from local emission with stagnation (LES) processes is important, because sometimes air pollution is not constrained within the geographical limits of an individual country, and pollutants can be transported via long-range transboundary processes (Chang et al., 2000; Kitada et al., 1993; Ichikawa et al., 1998; Carmichael et al., 2002). It is therefore impossible to solve the problem of air pollutants for an individual country alone; rather, regional/sub-regional agreements on pollutant-emission abatement strategies are required. These agreements would require quantitative estimates of the contributions of both local and long-range pollutants to the overall pollution.

Northeast Asia is affected by the high levels of sulfur dioxide (SO_2) and nitrogen oxide (NO_x) emissions from China (Ohara et al., 2007; Streets et al., 2006), and the LRT of pollutant from source regions with high levels of emissions (i.e., from the industrial centers of central or eastern China) is still one of the most serious environmental problem. Projections based on planned development patterns indicate that significant potential increases in emissions may occur during the next 25–50 years in East Asia (ESCAP, 2000), and the recent tropospheric satellite observations have demonstrated that NO_x emissions in China have accelerated impressively since 2000 (Richter et al., 2005; Ohara et al., 2007). Therefore, the LRT air-pollution process will still become an increasingly important environmental issue in northeast Asia.

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A number of air-quality models have been developed to study the transport and transformation mechanism of acidifying species, and dry and wet acid deposition, as well as the source–receptor relationship in East Asia (Wang et al., 2002; An et al., 2002; Park and Lee, 2003; Han et al., 2005; Kim et al., 2011). Although such modeling studies can now be performed for many pollutants, little fundamental researches on LRT indicators that can be more widely deployed and used more easily for the interpretation of LRT over northeast Asia, have been carried out over the last few decades. Indicators of LRT potential are to distinguish from others and quantify the degree of LRT contribution indirectly by investigating the relevant characteristic variables. Indicator studies have been generally carried out for ozone concentrations in order to distinguish between NO_x limited and VOC limited ozone formation (Kirchner et al., 2001; Sillman and He, 1997). These studies were performed to determine the limiting factor of ozone formation using measurement results, and ozone-related indicators have been tested in many modeling studies for many different locations.

However, few studies of either measurement or modeling have been carried out with other pollutants such as sulfur, nitrogen compounds, and particulate matters. Particulate matter, which is both a primary and a secondary air pollutant, is attracting considerable attention mainly due to its adverse impact on climate change and human health. It also helps control the concentration, lifetime, and physical and chemical behavior of many important tracer gases by providing reaction sites and by serving as a carrier and/or sink for many atmospheric species (Dentener et al., 1996; Rosenfeld et al., 2008). Therefore it is worthwhile investigating the simulated numerous chemical compounds comprehensively to substantiate LRT indicators of particulate matters over northeast Asia.

This paper proposes several indicators for distinguishing between LRT and LES cases for the days during which the concentration of PM_{10} concentration is high. LRT and LES differ both meteorologically and chemically. Modifiers such as transport, chemical transformations, and removal processes, which determine the endpoint concentrations, are observed from the sources of gas- and particulate-pollutant emission over northeast Asia. Although the particulate matter in this region originates from both natural and anthropogenic sources, in this paper, we will address the behaviors of anthropogenic emissions by employing a full modeling study using the MM5-CMAQ (Fifth-Generation Penn State/NCAR Mesoscale Model-Community Multi-scale Air Quality) modeling system. Several high-priority air pollutants or pollutant classes, such as gas- and particulate-phase air pollutants including NO_x, organic aerosol, and VOC-related species, are explored in order to determine the key indicators of LRT potential that can be used effectively as a general indicator in the interpretation of LRT over northeast Asia.

2. Method and data used

2.1. Derivation of indicators of LRT potential

As a first step in the quantification of the indicators of LRT potential, we first selected the days during which the concentration of particulate pollutants was high in Seoul and Busan (Fig. 1). We used air-quality data collected by the Korean Ministry of Environment from the year 2000–2007. We selected the days during which the haze phenomena occurred and at the same time concentration of observed PM_{10} was high with the daily mean exceeding 60 $\mu g\ m^{-3}$ in the urban cities of both Seoul and Busan, as indicated in Fig. 1.

Next, we classified high PM_{10} days into two extreme synoptic cases: LRT, and LES cases, and the representative days for LRT and LES were modeled and compared, and various air pollutants, pollutant classes, and ratios of chosen chemical species were explored as indicators of LRT potential. Therefore, in the current study, separating and distinguishing between LRT and LES-dominant cases is of considerable importance. Weather classification may be desirable, but is seldom feasible because there are many different synoptic weather patterns and a large number of variables that correspond to each case. The numerous methods for the classification of synoptic meteorological conditions were introduced and employed so far; such as cluster analysis (Davis and Gay, 1993), empirical orthogonal function analysis (Lorenz, 1956; Ludwig et al., 2002), Jenkinson Daily Weather Types (Lamb, 1972; Hulme and Barrow, 1997), and other empirical approaches (Buchholz et al., 2010). All these previous approaches are related to mean air pollution spatial patterns, and are not connected to tracking the moving (non-stagnant) air masses in association with the long-range transport process over a given sub-regional or meso-scale domain.

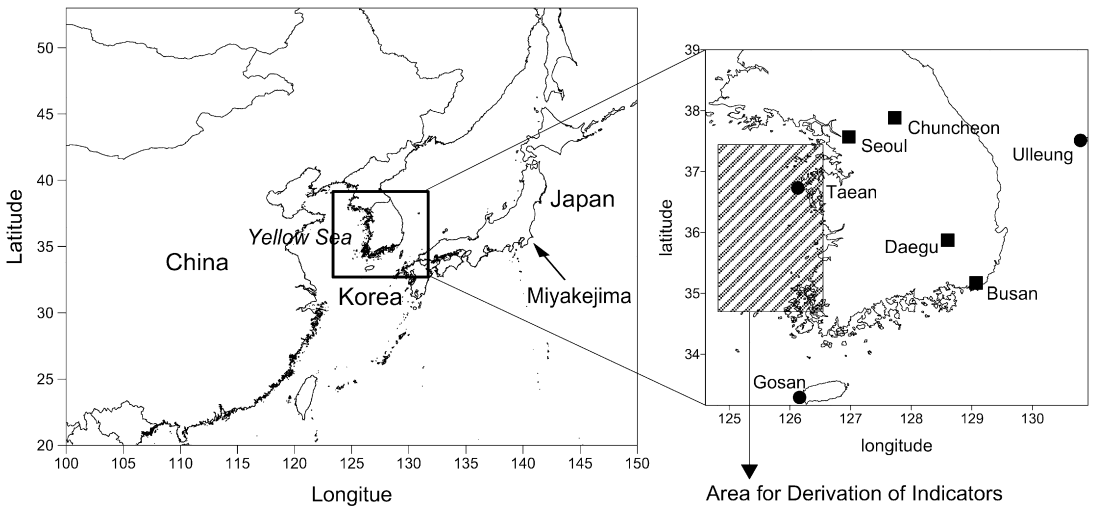

Fig. 1. Model domain, specific derivation area of LRT indicators, and locations of monitoring sites used for the model verification in the current study. The symbols (■) and (●) represent the locations of urban and background sites, respectively.