Science of the Total Environment 643 (2018) 692–703

Contents lists available at ScienceDirect

Science of the Total Environment

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

Simulation on different response characteristics of aerosol particle number concentration and mass concentration to emission changes over mainland China

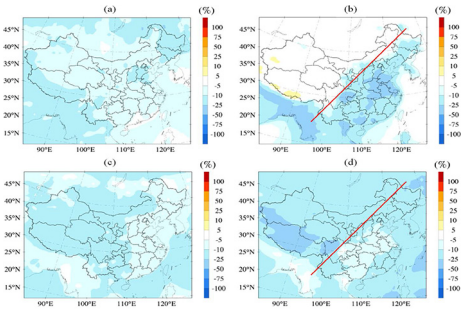
Xueshun Chen [a,b,*], Zifa Wang [a,b,d], Jie Li [a,b], Wenyi Yang [a,b], Huansheng Chen [a,b], Zhe Wang [a,c], Jianqi Hao [a], Baozhu Ge [a,b], Dawei Wang [a,b], Huili Huang [a]

[a] State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry (LAPC), Institute of Atmospheric Physics (IAP), Chinese Academy of Sciences (CAS), Beijing, China
[b] Center for Excellence in Urban Atmospheric Environment, Institute of Urban Environment, Chinese Academy of Sciences, Xiamen 361021, China
[c] Research Institute for Applied Mechanics (RIAM), Kyushu University, Fukuoka, Japan
[d] University of Chinese Academy of Sciences, Beijing 100049, China

HIGHLIGHTS

- PN concentration responds more heterogeneously to emission change than PM concentration.
- Both of primary particles and SO_2 emission reduction can decrease PM concentration.
- Reducing primary particles emission is effective in reducing PN concentration over eastern China.
- Reducing SO_2 emission is effective in reducing PN concentration over western China.
- Spatial difference of response of PN concentration to emission is distinguished by a boundary line.

GRAPHICAL ABSTRACT



1. Particle number concentration (Fig.b and Fig.d) responds more heterogeneously to emission change than particle mass concentration (Fig.a and Fig.c).
2. Reducing primary particles emission is more effective in reducing particles number concentration over central-eastern China (Fig.b) whereas reducing SO_2 emission is more effective over northwestern China (Fig.d).
3. 'Hu line' (labeled in red) is a also boundary line distinguishing responding characteristics of particles number concentration to emission change over different areas.

ARTICLE INFO

Article history:
Received 12 April 2018
Received in revised form 13 June 2018
Accepted 14 June 2018
Available online xxxx

Editor: Jianmin Chen

Keywords:
NAQPMS+APM
Aerosol microphysics
Number concentration
Mass concentration
Emission change

ABSTRACT

In this study, Nested Air Quality Prediction Modeling System with Advance Particle Microphysics module (NAQPMS+APM) is applied to simulate the response characteristics of aerosol particle number concentration and mass concentration to emission changes over mainland China. It is the first attempt to investigate the response of both aerosol mass concentration and number concentration to emission changes using a chemical transport model with detailed aerosol microphysics over mainland China. Results indicate that the response characteristics are obviously different between aerosol particle number concentration and mass concentration. Generally, the response of number concentration shows a more heterogeneous spatial distribution than that of mass concentration. Furthermore, number concentration has a higher sensitivity not only to primary particles emission but also to precursor gases than that of mass concentration. Aerosol particle mass concentration exhibits a consistent trend with the emission change and yet aerosol number concentration does not. Due to the nonlinearity of aerosol microphysical processes, reduction of primary particles emission does not necessarily lead to an obvious decrease of aerosol number concentration and it even increases the aerosol number

* Corresponding author at: State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry (LAPC), Institute of Atmospheric Physics (IAP), Chinese Academy of Sciences (CAS), Beijing, China.
E-mail address: chenxsh@mail.iap.ac.cn (X. Chen).

https://doi.org/10.1016/j.scitotenv.2018.06.181

concentration. Over Central-Eastern China (CEC), the most polluted regions in China, reducing primary particles emission rather than precursor gas emissions is more effective in reducing particles number concentration. By contrast, the opposite is true over the northwestern China. The features of fine particles pollution revealed in this study are associated with the spatial differences in China's population, geography, climate and economy. Considering the more adverse effects of ultrafine particles on human health and the spatial distribution of population, making different measures in controlling particles number concentration from that controlling mass concentration in different regions over mainland China is indicated.

Main findings: FPN concentration responds more heterogeneously to emission than FPM. Spatial difference of response of FPN to emission is distinguished by a boundary line.

1. Introduction

Over the past decades, the atmospheric composition of troposphere has been changed a lot due to anthropogenic activities (Akimoto, 2003; Tsigaridis et al., 2006). The increased atmospheric pollutants, such as aerosol particles have drawn much concern due to their important climate forcing (Charlson et al., 1992; Albrecht, 1989; Twomey, 1974, 1977), environmental and health effects (Han et al., 2012; Donaldson et al., 1998, 2002; Delfino et al., 2005) in global and regional scales. Evaluation and mitigation of these effects is an important issue for scientific community. The effects of aerosols depend on their physical and chemical properties, such as mass concentration, chemical composition, mixing state and number size distribution (Dusek et al., 2006; Oshima et al., 2009; Jacobson, 2014; Kulmala et al., 2013). It is necessary to understand how the aerosol physical and chemical properties respond to emission changes in order to take effective measure to reduce the adverse effects and project the influence of aerosols in the future (IPCC, 2007).

As we know, aerosols can be from primary emission and secondary formation (Seinfeld and Pandis, 1989). Primary particles emission increases both particle mass and number concentration. Secondary formation includes processes like nucleation from precursor gases, condensation process of condensable gases onto pre-existing particles and chemical processes. Nucleation increases both mass and number concentration while condensation and chemical process only increase mass concentration directly although they influence number concentration by altering particle size distribution. Several studies (Adams and Seinfeld, 2003; Stier et al., 2006; Reddington et al., 2011) have pointed out the importance of separating these two sources because they respond in different ways to gas and particle emission control strategies and environmental changes (Spracklen et al., 2006; Yu et al., 2012). Due to these reasons, it is expected that aerosol particle number concentration and mass concentration respond differently to emission control measures.

Currently, air quality legislation on particulate matter is mainly focused on mass concentration of fine particles with diameter <2.5 μm ($PM_{2.5}$) in most countries and regions (Kumar et al., 2010; Kumar et al., 2011). Fine particles mass (FPM) concentration is dominated by particles with diameter larger than 0.1 μm while fine particle number (FPN) concentration is dominated by ultra-fine particles (UPFs) with diameter smaller than 0.1 μm (Harrison et al., 2000; Wu et al., 2008). FPM concentration is not well associated with FPN concentration (Wehner et al., 2004) due to the different formation mechanisms of ultra-fine and larger particles (Seinfeld and Pandis, 1989). For combustion sources, some emission control technologies affect mainly the larger particle emissions, but they may even increase emissions of ultra-fine particles (Kumar et al., 2014). In addition, increase of new particle formation can supplement the loss of FPN and even increase the FPN concentration when reducing primary particles emission (Spracklen et al., 2006). Hence, in order to comprehensively estimate and project health and climate effects of anthropogenic aerosol particles, it is necessary to quantify the variations of FPM and FPN concentration with changes of emissions.

In China, along with the rapid economic development, atmospheric pollutant emissions have been remarkably increased in the last decades (Lei et al., 2011; Lu et al., 2011). As a result, the concentrations of critical pollutants, such as SO_2, NO_2, CO, O_3, $PM_{2.5}$ and PM_{10}, often exceed the regulatory limits by orders of magnitude (Zhang et al., 2013; Sharma et al., 2013). In this context, the main focus of air quality research is on FPM concentration and other critical pollutants, which are regulated by environmental protection administration. X. Wang et al. (2013) found that reduction of SO_2 emission from 2006 to 2009 decreased the concentration of SO_2 by 39% and sulfate by 26% in the Pearl River Delta region, China. Xue et al. (2013) reported that the emissions reduction of SO_2 and NO_x can decrease concentration of $PM_{2.5}$ by 2.23% in all cities in 2015 from the levels of 2010. The simulation of Y. Wang et al. (2013) indicated that total concentration of sulfate-nitrate-ammonium responds nonlinearly to emission changes of SO_2, NO_x, and NH_3. Saikawa et al. (2011) investigated the impact of vehicles emissions on the concentrations of O_3 and $PM_{2.5}$ over China. However, since there are no air quality regulations for UFPs, they have not been received due attention of regulatory authorities (Kumar et al., 2011) although they constitute the most contribution to FPN concentration. In addition, modeling FPN concentration requires a model to simulate particle's diameter down to nanometers with detailed microphysical processes, which is beyond the ability of models used by many previous studies on aerosol particle simulation over China (Luo and Yu, 2011). Although several advanced models have been developed to resolve aerosol microphysical processes and simulate particle number concentration in global and regional scales (Spracklen et al., 2008; Pierce and Adams, 2009; Zhou et al., 2012; Matsui et al., 2014), they are not widely used and evaluated comprehensively in China. There is a lack of attention on FPN concentration and how they respond to emission changes. Focus on both FPM concentration and FPN concentration is even fewer in China.

Considering the above, we investigate the responding behavior of fine particle mass concentration and number concentration simultaneously by using a regional chemical transport model with an advanced aerosol microphysics module covering diameter from 1 nm to 10 μm. The research results of our study are meaningful to understand fine particle pollution comprehensively over mainland China. Also, they can be used to provide policy suggestions for controlling fine particle pollution in terms of fine particle number concentration besides mass concentration. The NAQPMS+APM model is described in Sect. 2. The model configuration, input, and experiments setup are introduced in Sect. 3. The different responses of fine particle mass and number concentration to emission changes are discussed in Sect. 4. The conclusions are presented in Sect. 5.

2. Model description

NAQPMS+APM is a 3-D regional atmospheric chemical transport model with detailed aerosol microphysics to describe regional and urban scale atmospheric pollution. NAQPMS+APM has been used to study the spatial distribution of aerosol number concentration (Chen et al., 2014), evolution of particle number size distribution (Chen