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# Atmospheric Environment

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## Natural emissions under future climate condition and their effects on surface ozone in the Yangtze River Delta region, China



Min Xie <sup>a, b, \*</sup>, Lei Shu <sup>a</sup>, Ti-jian Wang <sup>a, b, c, \*\*</sup>, Qian Liu <sup>d</sup>, Da Gao <sup>a</sup>, Shu Li <sup>a</sup>,  
Bing-liang Zhuang <sup>a, b</sup>, Yong Han <sup>a</sup>, Meng-meng Li <sup>a</sup>, Pu-long Chen <sup>a</sup>

<sup>a</sup> School of Atmospheric Sciences, Nanjing University, Nanjing 210023, China

<sup>b</sup> Jiangsu Collaborative Innovation Center for Climate Change, Nanjing 210023, China

<sup>c</sup> CMA-NJU Joint Laboratory for Climate Prediction Studies, Institute for Climate and Global Change Research, School of Atmospheric Sciences, Nanjing University, Nanjing 210023, China

<sup>d</sup> Jiangsu Provincial Academy of Environmental Science, Nanjing 210036, China

### HIGHLIGHTS

- The natural emissions of ozone precursors are sensitive to climate in YRD.
- BVOCs and soil NO emissions will increase by 25.5% and 11.5% in the future.
- Future natural emissions cause about 20% of the surface O<sub>3</sub> increases, with the maximum of 2.4 ppb.
- O<sub>3</sub> formation in YRD will be insensitive to VOCs in the future.

### ARTICLE INFO

#### Article history:

Received 6 October 2016

Received in revised form

20 November 2016

Accepted 21 November 2016

Available online 24 November 2016

#### Keywords:

Climate change

Ozone

Natural emission

Indicator

The Yangtze River Delta region

### ABSTRACT

The natural emissions of ozone precursors (NO<sub>x</sub> and VOCs) are sensitive to climate. Future climate change can impact O<sub>3</sub> concentrations by perturbing these emissions. To better estimate the variation of natural emissions under different climate conditions and understand its effect on surface O<sub>3</sub>, we model the present and the future air quality over the Yangtze River Delta (YRD) region by running different simulations with the aid of the WRF-CALGRID model system that contains a natural emission module. Firstly, we estimate the natural emissions at present and in IPCC A1B scenario. The results show that biogenic VOC emission and soil NO<sub>x</sub> emission over YRD in 2008 is 657 Gg C and 19.1 Gg N, respectively. According to climate change, these emissions in 2050 will increase by 25.5% and 11.5%, respectively. Secondly, the effects of future natural emissions and meteorology on surface O<sub>3</sub> are investigated and compared. It is found that the variations in meteorological fields can significantly alter the spatial distribution of O<sub>3</sub> over YRD, with the increases of 5–15 ppb in the north and the decreases of –5 to –15 ppb in the south. However, only approximately 20% of the surface O<sub>3</sub> increases caused by climate change can be attributed to the natural emissions, with the highest increment up to 2.4 ppb. Finally, Ra (the ratio of impacts from NO<sub>x</sub> and VOCs on O<sub>3</sub> formation) and H<sub>2</sub>O<sub>2</sub>/HNO<sub>3</sub> (the ratio between the concentrations of H<sub>2</sub>O<sub>2</sub> and HNO<sub>3</sub>) are applied to study the O<sub>3</sub> sensitivity in YRD. The results show that the transition value of H<sub>2</sub>O<sub>2</sub>/HNO<sub>3</sub> will turn from 0.3 to 0.5 in 2008 to 0.4–0.8 in 2050. O<sub>3</sub> formation in the YRD region will be insensitive to VOCs under future climate condition, implying more NO<sub>x</sub> need to be cut down. Our findings can help us understand O<sub>3</sub> variation trend and put forward the reasonable and effective pollution control policies in these famous polluted areas.

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\* Corresponding author. School of Atmospheric Sciences, Nanjing University, Nanjing 210023, China.

\*\* Corresponding author. School of Atmospheric Sciences, Nanjing University, Nanjing 210023, China.

E-mail addresses: [minxie@nju.edu.cn](mailto:minxie@nju.edu.cn) (M. Xie), [tjwang@nju.edu.cn](mailto:tjwang@nju.edu.cn) (T.-j. Wang).

### 1. Introduction

Air quality deterioration and climate change are two critical challenges that concern the sustainable development of human society. Though they are separated in scientific research and

political considerations, many issues of them are closely related. For one thing, trace gases and aerosols have a direct or indirect effect on radiative forcing, which can lead to global warming or the changes of atmospheric circulation and hydrological cycles (Ramanathan et al., 2001; Karl and Trenberth, 2003; Forster et al., 2007; von Schneidemesser et al., 2015). For another, climate change can cause variations in meteorology, and thereby affect emission rates, transport conditions, chemical reactions and deposition processes of air pollutants (Isaksen et al., 2009; Jacob and Winner, 2009; von Schneidemesser et al., 2015). As the world enters an era of rapid global warming, climate change and its effect on air quality should be thoroughly investigated, in order to better protect our ambient environment as well as adapt to the changing climate (Jacob and Winner, 2009).

Ozone ( $O_3$ ) is one of the common air pollutants in troposphere, with adverse influence on human health and ecological balance (Fann and Risley, 2013; Landry et al., 2013; Xie et al., 2016b). Formed by a series of complex photochemical reactions between nitrogen oxides ( $NO_x = NO + NO_2$ ) and volatile organic compounds (VOCs),  $O_3$  are sensitive to the emissions of these precursors (Sillman, 1999; Xie et al., 2014) and the favorable meteorological conditions (Wise and Comrie, 2005; Leibensperger et al., 2008; Guo et al., 2009; Xie et al., 2016b). In the past decades, many modeling studies have investigated the future  $O_3$  concentrations under different climate scenarios to quantify the changes in  $O_3$  and clarify the potential drivers (Hogrefe et al., 2004; Langner et al., 2005; Dentener et al., 2006; Liao et al., 2006; Forkel and Knoche, 2007; Grewe, 2007; Meleux et al., 2007; Tagaris et al., 2007; Jiang et al., 2008; Lin et al., 2008; Nolte et al., 2008; Wu et al., 2008; Athanassiadou et al., 2010; Carvalho et al., 2010; Langner et al., 2012; Coleman et al., 2013; Doherty et al., 2013; Liu et al., 2013b; Young et al., 2013; Wang et al., 2013; Pfister et al., 2014). These researchers have found out that climate change can impact global and regional  $O_3$  in several ways, including changing meteorological factors, disturbing stratosphere-troposphere exchange, and enhancing natural emissions of ozone precursors (Jacob and Winner, 2009; von Schneidemesser et al., 2015).

In global and regional scale, biogenic VOCs (BVOCs) from vegetations and NO from soils are two important natural sources for  $O_3$  precursors. They are strongly dependent on various environmental factors including air temperature, solar radiation, leaf area, vegetation type, and some atmospheric species etc. (Williams et al., 1992; Stohl et al., 1996; Simpson et al., 1999; Guenther et al., 1995, 2006; Wang et al., 2005, 2007; Xie et al., 2007). These factors are likely to vary under different climate conditions, implying the natural emissions tend to change in the future (von Schneidemesser et al., 2015). Some previous investigations have predicted that natural isoprene emissions would increase by 30–80% in 2100 due to surface warming (Lathiere et al., 2005; Guenther et al., 2006; Wiedinmyer et al., 2006). Some found that future changes of land-use may affect BVOCs emissions to a lesser extent, with a great variation on different scenarios (Guenther et al., 2006; Lathiere et al., 2006; Wiedinmyer et al., 2006; Ashworth et al., 2013). Others thought that  $CO_2$  may impact the estimates of BVOCs emissions as well, but the exact role of  $CO_2$  in the future is not clearly clarified (Tai et al., 2013; Sun et al., 2013; von Schneidemesser et al., 2015). Till now, the characteristics and the magnitude of these nature emissions, as well as their effects on  $O_3$  formation, under the future climate scenarios are still uncertain. It is worthy to mention that investigations for the effects of future soil NO are quite limited. Thus, more relevant studies need to be carried out to improve our understanding.

In China, it was reported that climate change can cause  $O_3$  to increase in the east and decrease in the west from 2000 to 2050, and 40% of the increase may result from the strengthened BVOCs

emissions (Wang et al., 2013). For South China, climate change with the incidental increases in biogenic emissions can cause surface  $O_3$  to increase by 1.6 ppb (–5–5 ppb) in 2050, while the uncontrolled anthropogenic emissions will enhance the increment up to 12.8 ppb (Liu et al., 2013b). Obviously, the exact effects of climate change on  $O_3$  significantly vary in different regions. Being one of the world's most dynamic economic regions, the Yangtze River Delta (YRD) region in China has experienced severe  $O_3$  pollution as well (Chan and Yao, 2008; Ma et al., 2012; Ding et al., 2013; Xie et al., 2014, 2016a; 2016b; Zhu et al., 2015). Thus, it is necessary to quantify what extent the impact of future climate will be in YRD.

To fill the knowledge gap and offer a full understanding of the  $O_3$  variation trend over YRD, the air quality model system WRF-CALGRID is applied in this study to quantify the effects of climate change on  $O_3$  in this region of China. Considering that the future natural emissions and their effects on air quality have been seldom discussed for special purpose, we focus on and further discuss this issue. In Section 2, we describe the methodology and the input data adopted in this study. The model performance is evaluated in Section 3. In Section 4, future climate change and its impact on natural emissions are presented. In Section 5, the individual as well as the synthetic effects of future natural emissions and climate on  $O_3$  formation are investigated. In Section 6, sensitivities of surface  $O_3$ , as well as how to consider the effects of climate change in implementing emission reduction policies, are discussed. In the end, the conclusions and future research suggestions are given in Section 7.

## 2. Methodology

### 2.1. Model description

The three-dimensional air quality model system WRF-CALGRID, which consists of WRF and CALGRID, is adopted in this study. WRF is a new generation of meso-scale meteorology model, which is used to provide the off-line meteorological data for the chemical transport model CALGRID. Many previous studies have showed that WRF performs very well in all kinds of weather simulations and has a bright application prospect in providing the meteorological characteristics of air pollution in YRD (Wang et al., 2009; Xie et al., 2014, 2016a; 2016b; Liao et al., 2015; Zhu et al., 2015). With respect to CALGRID, it has been improved based on the original version that only contains the gas-phase chemistry (Xie et al., 2014). Up to now, the modified CALGRID is incorporated with a thermodynamic equilibrium model ISORROPIA (Nenes et al., 1998), a secondary organic aerosol model SORGAM (Schell et al., 2001) and a simplified aqueous-phase chemistry (Xie et al., 2014). The previous applications prove that it can efficaciously simulate the variations of primary pollutants (eg.,  $CH_4$ ,  $CO$ ,  $SO_2$ ,  $NO_2$ ,  $PM_{10}$ , and black carbon etc.), the amount of dry and wet deposition, and the formation of  $O_3$  and aerosols (Xie et al., 2007, 2009; 2014).

### 2.2. Basic configurations

Fig. 1 shows two nested domains that are used in both WRF and CALGRID. The outermost domain (Domain 1) covers the land areas of China with the center point at (36.3°N, 102.2°E), horizontal grids of  $94 \times 80$ , and grid spacing of 54 km. The nested domain (Domain 2) covers the eastern part of China with the center point at (31.9°N, 118.4°E),  $60 \times 78$  horizontal grids, and 18 km grid spacing. The red line highlights the areas of Shanghai, Jiangsu and Zhejiang, which is the core areas of the YRD region.

From the ground level to the top pressure of 100 hPa, there are 31 vertical sigma layers in WRF, with about 10 in the planetary boundary layer (PBL). The most important physics and dynamics

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