Biogenic isoprene emissions over China: sensitivity to the CO$_2$ inhibition effect

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ABSTRACT

Isoprene emissions emitted from vegetation are one of the most important precursors for tropospheric ozone and secondary organic aerosol formation. The authors estimate the biogenic isoprene emissions in China over 2006–2011 using a global chemical transport model (GEOS-Chem) driven by meteorological fields from the assimilated meteorological data from MERRA. The authors incorporate three different parameterizations of isoprene–CO$_2$ interaction into the model, and perform three sensitivity simulations to investigate the effect of CO$_2$ inhibition on isoprene emissions for the period 2006–2011 in China. The annual isoprene emissions rate across China is simulated to be 12.62 Tg C yr$^{-1}$, averaged over 2006–2011, and decreases by about 2.7%–7.4% when the CO$_2$ inhibition schemes are included. The CO$_2$ inhibition effect might be significant in regions where the CO$_2$ concentration and isoprene emissions are high. Estimates of isoprene emissions can differ depending on the scheme of CO$_2$ inhibition. According to the results obtained from the sensitivity simulations, the authors find that the CO$_2$ inhibition effect leads to 5.6% ± 2.3% reductions in annual isoprene emissions over China. The authors also find that inclusion of CO$_2$ inhibition can substantially alter the sensitivity of isoprene emissions to the changes in meteorological conditions during the study period.

1. Introduction

Isoprene is a volatile organic compound (VOC) mainly emitted from terrestrial vegetation, and it makes up the largest fraction of non-methane biogenic VOCs, with an estimated emissions rate of 400–600 Tg C yr$^{-1}$ at the global scale (Guenther et al. 2006; Arneth et al. 2008). In polluted regions, biogenic isoprene emissions are an important contributor to tropospheric ozone formation in the presence of nitrogen oxides (NO$_x$), but in remote regions with low-NO$_x$ concentration, isoprene could reduce ozone by sequestering NO$_x$ as isoprene nitrate or by ozonolysis (Fiore et al. 2012). In addition, isoprene acts as a major precursor for secondary organic aerosol (SOA) formation, and can affect the atmospheric oxidation capacity through influencing the regional level of tropospheric hydroxyl radicals (OH) and the lifetime of methane (Peñuelas and Staudt 2010). Therefore, changes in isoprene emissions could modulate atmospheric composition and chemistry. An accurate estimate of isoprene emissions is important for air quality and climate change studies, and thus warrants in-depth investigation.

Many previous studies have shown that biogenic isoprene emissions are not only dependent on changes in environmental factors, such as canopy temperature, light, soil moisture etc., but also related to changes in vegetation type, vegetation distribution, leaf area, and leaf age (Guenther et al. 2006). Some recent studies have reported that changes in atmospheric CO$_2$ concentration might...
promote or limit isoprene emissions from vegetation. Increasing CO\textsubscript{2} concentration could enhance vegetation productivity (Piao et al. 2011), and hence indirectly promote isoprene emissions. However, it is unclear whether a raised atmospheric CO\textsubscript{2} concentration would increase isoprene emissions intrinsically (Penuelas and Staudt 2010). Several laboratory and field studies have indicated that the isoprene emissions rate has an inverse relationship in response to rising CO\textsubscript{2} concentration in the short and long term because an elevated CO\textsubscript{2} concentration might uncouple isoprene emissions from photosynthesis and suppress isoprene emissions at leaf level (Rosenstiel et al. 2003; Possell, Hewitt, and Beerling 2005) (known as ‘the CO\textsubscript{2}-inhibition effect’).

A number of previous studies have attempted to introduce the CO\textsubscript{2}-inhibition effect into chemical transport models for examining the impact of climate change on isoprene emissions, although the relationship between CO\textsubscript{2} and isoprene is not fully understood (Arnth et al. 2007; Heald et al. 2009; Wilkinson et al. 2009; Lathière, Hewitt, and Beerling 2010; Possell and Hewitt 2010). Arneth et al. (2007) found that observed leaf isoprene emissions were reproduced well by implementing the isoprene response to CO\textsubscript{2} concentration into the model used in their study, which is expressed as the ratio of the leaf internal CO\textsubscript{2} concentration at ambient CO\textsubscript{2} = 370 ppmv to the leaf internal CO\textsubscript{2} concentration. They also suggested the CO\textsubscript{2}-inhibition effect could be large enough to counteract the increases in isoprene emissions due to CO\textsubscript{2}-induced enhancement of vegetation productivity and leaf area growth. According to the isoprene measurements taken from aspen trees growing under four different CO\textsubscript{2} concentrations, Wilkinson et al. (2009) proposed a sigmoidal, Hill-reaction type isoprene–CO\textsubscript{2} curve to describe the short-term and long-term isoprene response to changes in atmospheric CO\textsubscript{2}. Heald et al. (2009) used a global coupled land–atmosphere model with the CO\textsubscript{2}-isoprene parameterization of Wilkinson et al. (2009) to explore the potential role of CO\textsubscript{2} in isoprene emissions over 2000–2100. They suggested the projected increases in isoprene emissions due to the warming climate in 2100 could be significantly modified by including the CO\textsubscript{2} inhibition effect. Recently, Possell and Hewitt (2010) improved the isoprene–CO\textsubscript{2} response curve by considering a wide range of tree species from tropical to temperate regions. The aforementioned studies indicate the important impacts of changes in atmospheric CO\textsubscript{2} concentration on isoprene emissions simulation, but large discrepancies remain among these isoprene–CO\textsubscript{2} relationships and related parameters. Such differences can result in different isoprene emissions predictions. Recently, a number of studies have examined biogenic emissions in China (Li et al. 2012; Li, Chen, and Xie 2013; Fu and Liao 2014; Li and Xie 2014). However, those studies were mostly focused on the estimation and spatiotemporal variation of biogenic VOC emissions, and investigating the roles of meteorological factors and vegetation parameters in biogenic emissions. No previous studies have quantified the impact of CO\textsubscript{2} concentration on isoprene emissions in China, or evaluated the uncertainty of the CO\textsubscript{2}-inhibition effect.

In this study, we use a global chemical transport model (GEOS-Chem) to estimate the biogenic isoprene emissions in China over 2006–2011, and examine the effect of CO\textsubscript{2} inhibition on regional isoprene emissions. We quantify the CO\textsubscript{2}-inhibition effect on the simulation of isoprene emissions and the uncertainty in comparison with different CO\textsubscript{2} inhibition parameterizations in the model, based on previous studies. We further discuss the implications for regional air quality due to the inclusion of CO\textsubscript{2} inhibition effects on isoprene emissions.

2. Model and methods

We use the GEOS-Chem global 3D chemical transport model, version 9-02 (http://acmg.seas.harvard.edu/geos/) to simulate the biogenic isoprene emissions in China over 2006–2011. The model is driven by the assimilated meteorological data from MERRA (http://gmao.gsfc.nasa.gov/merra/), with a horizontal resolution of 2.0º latitude × 2.5º longitude and a reduced vertical resolution of 47 levels. A similar modelling framework was used by Fu and Tai (2015). In GEOS-Chem, biogenic isoprene emissions are calculated by the Model of Emissions of Gases and Aerosols from Nature (MEGAN v2.1) (Guenther et al. 2006, 2012), which is estimated as a function of plant functional type-specific emission factors (E\textsubscript{p}, \textmu g C m\textsuperscript{-2} h\textsuperscript{-1}) modulated by environmental activity factors (normalized ratio) to account for the effects of temperature (\textgamma\textsubscript{T}), light (\textgamma\textsubscript{L}), leaf age (\textgamma\textsubscript{age}), LAI, soil moisture (\textgamma\textsubscript{sm}) and CO\textsubscript{2} concentration (\textgamma\textsubscript{CO\textsubscript{2}}). The biogenic isoprene emissions rate (E) in each model grid cell is computed as

\[
E = E_0 \cdot \text{LAI} \cdot \textgamma\textsubscript{T} \cdot \textgamma\textsubscript{L} \cdot \textgamma\textsubscript{age} \cdot \textgamma\textsubscript{sm} \cdot \textgamma\textsubscript{CO\textsubscript{2}} .
\] (1)

However, the default model does not consider the effect of soil moisture and CO\textsubscript{2} inhibition by setting \textgamma\textsubscript{sm} = 1 and \textgamma\textsubscript{CO\textsubscript{2}} = 1. To account for the CO\textsubscript{2}-inhibition effect, the empirical relationships between CO\textsubscript{2} concentration and the isoprene emissions rate from previous studies are applied in this work. For examining the impact of CO\textsubscript{2} inhibition on isoprene simulation, we perform four sets of simulations: [noCO\textsubscript{2}\_ctrl], [wCO\textsubscript{2}\_A], [wCO\textsubscript{2}\_W], and [wCO\textsubscript{2}\_P]. For each set, a six-year simulation is performed with meteorological fields from 2006 to 2011, present-day vegetation parameters and fixed anthropogenic emissions at year-2005 levels (Streets et al. 2003; Zhang et al. 2009). The simulation [noCO\textsubscript{2}\_ctrl] is the control
simulation without the CO$_2$-inhibition effect. The simulations [wCO$_2$A], [wCO$_2$W], and [wCO$_2$P] are the sensitivity simulations, which are similar to [noCO$_2$_ctrl] but with different CO$_2$-inhibition parameterizations. In the simulation [wCO$_2$A], the empirical CO$_2$–isoprene relationship is from Arneth et al. (2007) (Equation (2)); and in the simulation [wCO$_2$W], the CO$_2$ inhibition parameterization of Wilkinson et al. (2009) is used (Equation (3)). The simulation [wCO$_2$P] applies the CO$_2$-inhibition effect given by Possell and Hewitt (2010) (Equation (4)). The CO$_2$ concentrations used for calculating the $\gamma_{CO_2}$ in all the simulations are from the GEOS-Chem CO$_2$ simulation during the same period.

As shown in Arneth et al. (2007), the additional activity factor associated with the CO$_2$ suppressed effect can be modelled in Equation (2):

$$\gamma_{CO_2} = \frac{C_{i-370}}{C_i}, \quad (2)$$

where $C_i$ is the leaf internal CO$_2$ concentration, and $C_{i-370}$ is the leaf internal concentration at ambient CO$_2$ = 370 ppmv (under non-water-stressed conditions). According to Possell, Hewitt, and Beerling (2005), $C_i$ is about 70% of the ambient CO$_2$ concentration ($C_i$).

We also apply the isoprene–CO$_2$ relationship from Wilkinson et al. (2009), which is

$$\gamma_{CO_2} = I_{max} \frac{I_{max} \times (C_i)^h}{(C_{i}^\alpha + (C_{i}^\beta)^h)}, \quad (3)$$

where $I$ is the isoprene emissions rate, $I_{max}$ is the estimated asymptote at which further decreases in CO$_2$ concentration ($C_i$) would suppress isoprene emissions, and $C_{i}$ and $h$ are the Hill-type coefficients used to adjust the sigmoidal slope of the relationship between $I_{max}$ and $C_i$. In this study, the $I_{max}$, $C_{i}$, and $h$ are determined from the measurements of plants grown at four different CO$_2$ concentrations (400, 600, 800, and 1200 ppmv), by best-fit lines. The parameters are obtained from Wilkinson et al. (2009, Table 1).

The third normalized ratio to account for the effect of CO$_2$ concentration is provided by Possell and Hewitt (2010),

$$\gamma_{CO_2} = \frac{a}{1 + a \times b \times C_i}, \quad (4)$$

where $\gamma_{CO_2}$ is 1 at a CO$_2$ concentration equal to 370 ppmv, and $a$ and $b$ are empirical coefficients. Here, we use the fitting parameters $a = 8.9406$ and $b = 0.0024$ ppm$^{-1}$, which are provided in Possell and Hewitt (2010, Figure 5).

### 3. Results

Without the CO$_2$ effect ([noCO$_2$_ctrl]), the simulated annual isoprene emissions rate averaged over 2006–2011 across China is about 12.62 Tg C yr$^{-1}$. The annual isoprene emissions rate simulated in this range is within the range of 9.3–23.4 Tg C yr$^{-1}$ reported for China (Fu and Liao 2012; Li, Chen, and Xie 2013). Isoprene emissions are highest in summer (June–July–August, JJA) and lowest in winter (December–January–February, DJF). The isoprene emissions in DJF, MAM (March–April–May), JJA, and SON (September–October–November) account for 4.8%, 18.5%, 55.0%, and 21.7% of the annual emissions, respectively (Table 1). Figure 1(a) shows the spatial distribution of summertime and annual mean isoprene emissions from the [noCO$_2$_ctrl] simulation averaged over 2006–2011. We find that, largely, isoprene emissions are simulated over southern (south of 35°N) and northeastern China in summer, which are within the range of 10–40 mg C m$^{-2}$ d$^{-1}$, and mostly attributable to the increases in temperature and vegetation density. In addition, the spatial distribution of isoprene emissions is generally consistent with the distribution of trees in China, as trees are considered the highest isoprene emitter, compared with other vegetation types such as crops and grass.

We find that the spatial patterns of CO$_2$ effects on isoprene emissions are similar across China, despite the amount of influence exhibiting some discrepancies among the three different CO$_2$-inhibition parameterizations (Figure 1(b–d)). As shown in Figure 1, the CO$_2$ effect can substantially reduce isoprene emissions in summer in most of eastern China, especially in the eastern regions of Sichuan Province and southeastern China. The strong reductions in isoprene emissions in those regions are primarily due to the atmospheric CO$_2$ concentrations in those regions being generally higher than in other regions. As reported

### Table 1. Estimates of isoprene emission rates in China averaged over 2006–2011 (Tg C yr$^{-1}$). Also shown are the percentage changes of isoprene emissions (%) between the experiments with ([wCO$_2$A], [wCO$_2$P], and [wCO$_2$W]) and without ([noCO$_2$_ctrl]) the CO$_2$-inhibition effect.

| Isoprene | [noCO$_2$_ctrl] | [wCO$_2$A] – [noCO$_2$_ctrl] | [wCO$_2$P] – [noCO$_2$_ctrl] | [wCO$_2$W] – [noCO$_2$_ctrl] |
|----------|-----------------|-----------------------------|-----------------------------|-----------------------------|
| Annual   | 12.62           | –7.4                        | –6.6                        | –2.7                        |
| Winter   | 0.60            | –7.9                        | –7.1                        | –2.7                        |
| Spring   | 2.34            | –8.3                        | –7.5                        | –2.7                        |
| Summer   | 6.93            | –7.1                        | –6.3                        | –2.6                        |
| Autumn   | 2.75            | –7.3                        | –6.6                        | –2.6                        |
Figure 1. (a) Simulated summertime (left column) and annual (right column) biogenic isoprene emissions averaged over 2006–2011 in China in [noCO₂_ctrl]. (b) Spatial distribution of changes in isoprene emissions as a result of the CO₂-inhibition effect using the scheme of Arneth et al. (2007) ([wCO₂_A] – [noCO₂_ctrl]). (c) As in (b) but with the scheme of Possell and Hewitt (2010) ([wCO₂_P] – [noCO₂_ctrl]). (d) As in (b) but with the scheme of Wilkinson et al. (2009) ([wCO₂_W] – [noCO₂_ctrl]).

by a number of laboratory-based studies, when CO₂ changes within the range of 200–1200 ppmv, trees grown at lower CO₂ concentrations exhibit significantly higher isoprene emission rates compared with those grown at
Figure 2 represents the effects of CO$_2$ inhibition on seasonal isoprene emissions over China during 2006–2011 from $[\text{wCO}_2\_A] - [\text{noCO}_2\_\text{ctrl}]$, $[\text{wCO}_2\_P] - [\text{noCO}_2\_\text{ctrl}]$, and $[\text{wCO}_2\_W] - [\text{noCO}_2\_\text{ctrl}]$. In all seasons, the maximum reduction in isoprene emissions due to the CO$_2$ effect is obtained in $[\text{wCO}_2\_A]$, followed by $[\text{wCO}_2\_P]$ and $[\text{wCO}_2\_W]$. The CO$_2$ effect on isoprene emissions exhibits little seasonal variation in all sensitivity simulations. However, the changes in isoprene emissions resulting from CO$_2$ inhibition display interannual variation during 2006–2011, except those in $[\text{wCO}_2\_W]$. In $[\text{wCO}_2\_A]$, the isoprene emissions in DJF over China decrease by $-7.8\%$ (median value) when taking into account CO$_2$ inhibition, and the decline in isoprene emissions in MAM due to CO$_2$ inhibition varies from $-9.8\%$ to $-10\%$.
−6.6%, with a median of −8.4%. In JJA and SON, the CO₂ effect leads to a decrease in isoprene emissions of −8.5% to −6.0% in [wCO₂_A] over 2006–2011. The reductions in isoprene emissions induced by the CO₂ effect in [wCO₂_P] are similar to the results of [wCO₂_A]. We also find that the interannual variation in isoprene emissions, induced by the effect of CO₂ inhibition, is quite important compared to the impact of land-cover and land-use change. As shown by Fu and Liao (2012), simulated isoprene emissions in summer over eastern China change by 5%–8% as a result of vegetation change alone over 2001–2006.

As shown above, estimates of isoprene emissions can differ depending on the CO₂–isoprene response curve, which also represents a major source of uncertainty in projecting future isoprene emissions as the atmospheric CO₂ concentration continues to rise. The discrepancies in the three CO₂–isoprene relationships likely result from the differences in quantitative algorithms and empirical coefficients, which are obtained from different plant species in growth-chamber experiments. For example, some studies describe the response as a purely mathematical relationship based on the experimental growth of two isoprene-emitting herbaceous species under different CO₂ levels (Possell, Hewitt, and Beerling 2005; Arneth et al. 2007). Whereas, Wilkinson et al. (2009) constructed an empirical relationship through consideration of the principles of enzyme kinetics based on the measured responses of temperate cottonwood and aspen trees under controlled-environment growth chambers. Possell and Hewitt (2010) attempted to define the CO₂-inhibition effect using laboratory measurements of tropical tree species (Acacia nigrescens). In order to better understand the calculated CO₂ inhibition in the model, we further quantify the CO₂-inhibition effect and its uncertainty according to the results of the sensitivity simulations. As shown in Figure 3, in the presence of CO₂–isoprene interaction, the annual present-day (2006–2011) isoprene emissions over China reduce by 5.6% ± 2.3%, while the isoprene emissions in DJF, MAM, JJA, and SON are cut by 5.9% ± 2.5%, 6.2% ± 2.7%, 5.3% ± 2.1%, and 5.5% ± 2.2%, respectively.

The significance of the variations induced by CO₂ inhibition can also be demonstrated when compared with the changes in isoprene emissions resulting from climate change alone. For instance, without the CO₂ effect, changes in meteorological conditions between the two three-year periods of 2006–2008 and 2009–2011 enhances summertime isoprene emissions by about 50 Gg C/month in China (1 Gg = 10⁹ g) (isoprene averaged over 2009–2011 minus isoprene averaged over 2006–2008). However, inclusion of the CO₂ effect can partly offset such increases or even reverse the sign. The simulated summertime isoprene increment from the period 2006–2008 to the period 2009–2011 on average shrinks by 20% when the CO₂ effect is considered in [wCO₂_W], while the CO₂ effect in [wCO₂_A] and [wCO₂_P] can completely nullify such an increase and lead to 70 Gg C/month and 60 Gg C/month reductions in isoprene emissions, respectively. The results in this study imply that the inclusion of CO₂ inhibition can substantially affect the sensitivity of isoprene emissions to changes in meteorological conditions. The impact of CO₂ inhibition can be more significant on multi-decadal scales than the magnitudes reported here. Recently, a few studies have indicated that the inclusion of CO₂ inhibition would generally reduce the sensitivity of air pollution to climate and vegetation change under future projection. Tai et al. (2013) reported that, over 2000–2050, the inclusion of CO₂ inhibition leads to reduced sensitivity of surface ozone and SOA (by more than 50%) to climate and natural vegetation change where isoprene emissions are important, implying a benefit of air quality in populated, high-NO₂ regions.

4. Discussion and conclusions

A global transport model (GEOS-Chem) is used in this study to simulate the isoprene emissions over China, with the inclusion of CO₂–isoprene interaction, from 2006 to 2011. Without the CO₂-inhibition effect, the simulated isoprene emissions rate is approximately 12.62 Tg C yr⁻¹ across China. To quantify the impact of CO₂ inhibition on isoprene emissions, three estimates of isoprene emissions with different parameterizations of the CO₂–isoprene response curves are compared. The results indicate that the CO₂-inhibition effect, which is not included in most chemistry or climate modelling studies, is significant in estimating isoprene emissions. For instance, applying the Wilkinson et al. (2009) scheme in [wCO₂_W] decreases annual isoprene emissions by −3% relative to the control simulation ([noCO₂_ctrl]).
without CO₂ inhibition. Whereas, applying the CO₂ inhibition scheme of Arneth et al. (2007) in [wCO₂ A] and Possell and Hewitt (2010) in [wCO₂ P] reduces annual isoprene emissions by ~7% over China. This effect might be significant in regions where the CO₂ concentration and isoprene emissions are high. To summarize, the impact of CO₂ inhibition can lead to an annual isoprene emissions decrease of 5.6% ± 2.3%. Regionally, summertime isoprene emissions might be cut by more than 9% when the CO₂-inhibition effect is included. Compared with the changes in isoprene emissions resulting from climate change alone on the multi-decadal scale, the reductions in isoprene emissions induced by CO₂ inhibition are significant. Sensitivity studies have shown that, in China, changes in meteorological conditions between the late 1980s and mid-2000s led to increases in isoprene emissions by 17% (Fu and Liao 2014). The changes in isoprene emissions resulting from climate change can be modified if the CO₂ inhibition is accounted for in the model.

There are a few studies that have indicated that the CO₂–isoprene effect might have a potential influence for projected ozone air quality or SOA concentrations under future climate change scenarios (Young et al. 2009; Tai et al. 2013), because they are both sensitive to the spatial and temporal variations of biogenic isoprene emissions (Fu and Liao 2012). In this study, the inclusion of CO₂ inhibition may lead to a reduction in SOA concentrations (by ~10%) where isoprene emissions largely decrease. Future work should focus on a more systematic analysis of the variation of in ozone and SOA to CO₂–isoprene integration under climate change. However, the CO₂–isoprene response curves are built on a limited number of measurements for several species in earlier studies, so the parameterizations of CO₂–isoprene interaction still pose a challenge for accurate estimates of isoprene emissions in China at present. In addition, a few previous experimental studies pointed out that inhibition of the isoprene emissions rate occurs in the presence of an increased CO₂ concentration for both short-term exposure (seconds to minutes) and long-term exposure (weeks to months). The responses of isoprene emissions to changes in CO₂ concentration might be different on various time scales. For instance, the response of isoprene emissions might be driven by adjustments in existing metabolic components during a single day. Whereas, on time scales at which leaves develop and grow (weeks or months), the response of isoprene emissions is likely driven by the adjustments in gene expression and the production of new metabolic components (Wilkinson et al. 2009). Here, we only focus on the effects of CO₂ inhibition on monthly and seasonal isoprene emissions, rather than diurnal isoprene emissions, mostly because the changes in sub-ambient CO₂ concentration (intercellular CO₂) over shorter time scales are scarce. The short-term effect of CO₂ inhibition on daily isoprene emissions is still a challenge and full of large uncertainty, especially in China. Wilkinson et al. (2009) reported that the sensitivity of the isoprene emissions rate to intercellular CO₂ could decrease with long-term exposure to increased atmospheric CO₂ if the intercellular CO₂ concentration changes between 200 and 400 ppmv. Since the diurnal variation of isoprene emissions is strong, the diurnal effect of CO₂ concentration on isoprene emissions definitely warrants further investigation. More specific information on, and measurements of, extensive and representative plant species from major isoprene-release regions are required to improve CO₂–isoprene parameterization in future studies in China.

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