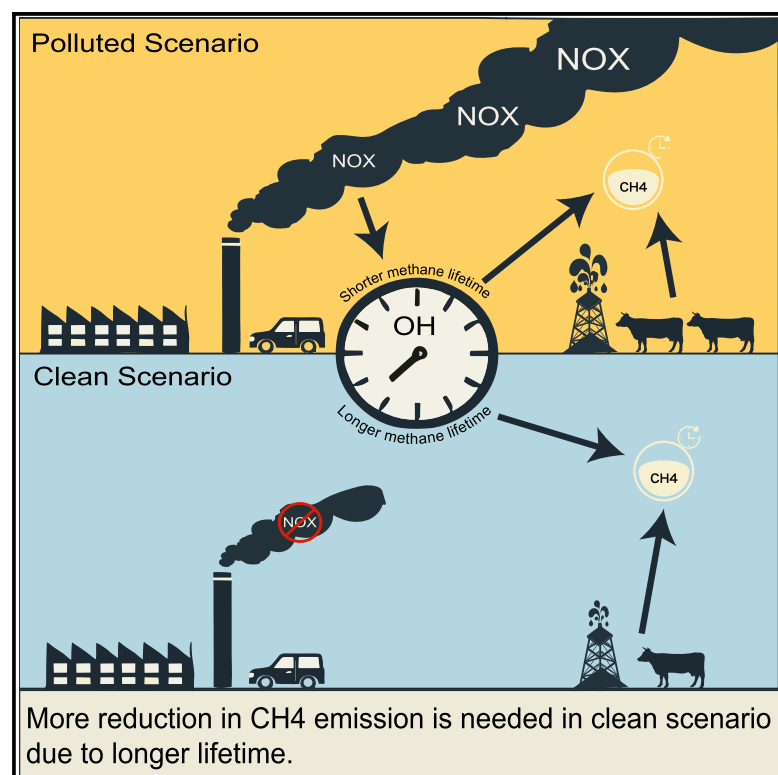


Clean air policy makes methane harder to control due to longer lifetime

Graphical abstract



Authors

Bo Fu, Jingyi Li, Yongye Jiang,
Zhiwei Chen, Bengang Li

Correspondence

libengang@pku.edu.cn

In brief

Tackling climate change and improving air quality are connected goals with hidden drawbacks. This study shows that reducing air pollutants like nitrogen oxides can make it harder to control methane. Policymakers must consider these interactions to avoid overly optimistic estimates and consider more reduction efforts.

Highlights

- The emissions of NO_x, CO, and VOCs significantly affect CH₄ lifetime
- NO_x emissions changes determine the overall effect of the three pollutants
- Reducing pollutants will extend CH₄ lifetime and make it harder to control



Article

Clean air policy makes methane harder to control due to longer lifetime

Bo Fu,¹ Jingyi Li,¹ Yongye Jiang,¹ Zhiwei Chen,¹ and Bengang Li^{1,2,3,*}¹College of Urban and Environmental Sciences, Institute of Carbon Neutrality, Peking University, Beijing 100871, China²Jiangsu Center for Collaborative Innovation in Geographical Information Resource Development and Application, Nanjing 210023, China³Lead contact

*Correspondence: libengang@pku.edu.cn

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SCIENCE FOR SOCIETY Tackling climate change and improving air quality are both essential goals. While working on one often helps the other, there can also be hidden drawbacks that we do not always consider. This article looks at one of these hidden issues: how cleaning up air pollution might make it harder to control methane (CH₄), a powerful greenhouse gas. CH₄ stays in the atmosphere for a certain amount of time, and this can be affected by air pollutants like nitrogen oxides. Reducing these pollutants is good for air quality but can extend the time CH₄ stays, making it harder to reduce CH₄ levels and fight climate change. Our study shows that under clean air scenarios, it is 20% harder to control CH₄ compared to moderate pollution scenarios. This means we would need to work even harder to reach our CH₄ reduction goals. These findings highlight the complexity of achieving clean air and climate targets together. Policymakers need to consider these interactions to avoid unexpected setbacks.

SUMMARY

Achieving the Paris Agreement's 1.5°C target necessitates reversing the rise in atmospheric concentrations of methane (CH₄), which is a greenhouse gas that is more radiatively potent than carbon dioxide, and that possesses a considerably shorter lifetime. Future reductions in pollutants like nitrogen oxides for air quality improvement are anticipated, with a side effect of potentially extending the lifetime of CH₄. However, at present the antagonism between air quality improvements and climate change response with respect to CH₄ lifetime is not being prominently addressed. Utilizing the GEOS-Chem model, we assessed CH₄ lifetime sensitivity to pollutant emissions. Applying this sensitivity to the OSCAR box model, we simulated future CH₄ dynamics, revealing that pollutant reduction in SSP1-26 compared to SSP2-45 could offset nearly 20% of CH₄ abatement efforts. Our study highlights the pollution abatement penalty in controlling atmospheric CH₄ concentrations, suggesting the need for escalated endeavors to combat climate change.

INTRODUCTION

Reducing methane (CH₄) emissions is recognized as important for mitigating climate change.^{1–3} Compared to carbon dioxide (CO₂), CH₄ has a higher global warming potential and a much shorter lifetime (~9 years). Currently, the CH₄ concentration has more than doubled compared to preindustrial levels, contributing to a radiative forcing of 0.54 W/m², second only to CO₂.⁴ Fortunately, reductions in CH₄ emissions can reduce its atmospheric concentration and climate impact in the short term.⁵ It is therefore believed that reducing CH₄ emissions will buy time for humanity to tackle climate change.⁶ Recently, international cooperation and national policies on CH₄ reduction have emerged, meaning that the importance of CH₄ control

has been recognized by the global community.^{7,8} The announcements of a national action plan on CH₄ was stated in the US-China Joint Glasgow Declaration on Enhancing Climate Action in the 2020s. In addition, at the UN Climate Change Conference (COP26) in 2021, more than 100 countries signed the Global Methane Pledge (GMP), committing to a 30% reduction in emissions by 2030 compared to 2020.⁹ These pledges offer a glimmer of hope for the Paris Agreement to achieve its goals.¹⁰

The atmospheric lifetime of CH₄ is sensitive to the hydroxyl radical (OH). OH breaks down CH₄ and is the main pathway via which CH₄ is removed from the atmosphere.^{11–13} Even a small change in OH concentrations can have a large impact on the atmospheric CH₄ concentration.^{11–15} Therefore, the extent to which reductions in CH₄ emissions can lead to reductions in

atmospheric CH₄ concentrations depends largely on the amount of OH in the atmosphere. OH concentrations are sensitive to air pollutants emissions. Nitrogen oxides (NO_x) promote the formation of OH, while the oxidation of carbon monoxide (CO) and non-CH₄ volatile organic compounds (VOCs) in the atmosphere consumes OH. Anthropogenic emissions of the above three ozone precursor pollutants (NO_x, CO, and VOCs) significantly modulate the lifetime and concentration of CH₄.^{16–20} Reductions in these pollutants (with NO_x dominating the total effect) have accelerated CH₄ concentration growth, as observed during the COVID-19 pandemic lockdown period.^{11,14,21–23} Although the reduction in pollutant emissions during the pandemic was passive due to the lockdown, it provided a glimpse of the future clean air scenario.²² As pollutant emissions are reduced, a longer lifetime of CH₄ can be expected, making it more difficult to reduce atmospheric CH₄ abundance. However, the impact of pollutant removal on the difficulty of CH₄ control is not well understood and has received little attention, potentially challenging the achievement of the Paris Agreement targets.

In this study, we designed a series of scenarios for pollutant emissions reductions and CH₄ mitigation efforts. Our findings indicate that reducing pollutants such as NO_x significantly extends the lifetime of CH₄, thereby diminishing the effectiveness of CH₄ concentration control initiatives like the GMP. Achieving the same CH₄ concentration control under clean scenarios requires more intensive CH₄ reduction efforts compared to moderate and polluted scenarios. These results underscore the impact of air pollution abatement penalties on the lifetime and effectiveness of CH₄ mitigation efforts. This result highlights the need for both scientific and societal attention to focus more on the antagonistic effects between climate change mitigation and air quality control, rather than just the synergistic effects, to avoid overly optimistic expectations and insufficient reduction efforts.

RESULTS

Methods summary

Here, we created multiple scenarios based on Shared Socioeconomic Pathways (SSPs)²⁴ and the GMP. Initially, we conducted a series of sensitivity experiments using GEOS-Chem,²⁵ a three-dimensional (3D) chemical transport model, to simulate CH₄ lifetimes under varying pollutant emissions. Subsequently, we quantified the sensitivity of CH₄ lifetime to pollutant emissions through regression analysis based on GEOS-Chem results. By applying this sensitivity to the OSCAR²⁶ (Outcome, Situation, Choices, Actions, and Review) model, a box model, we simulated atmospheric CH₄ concentrations in different scenarios and compared the effectiveness of CH₄ reduction under various conditions.

The effect of three pollutants on CH₄ lifetime

Initially, we performed a series of sensitivity experiments by varying emissions of NO_x, CO, and VOCs using GEOS-Chem to quantify the impact of emissions on CH₄ lifetime. Fifty experiments were carried out under identical meteorological conditions and CH₄ concentrations, differing only in pollutant emissions and consequently affecting CH₄ lifetime (see Table S1). The results show that a 50% reduction in present anthropogenic CO or VOCs can result in an about 3% decrease in CH₄ lifetime.

In comparison, a 50% reduction in NO_x emissions could result in an 8% increase in CH₄ lifetime (Figures 1A and 1B). Reductions in air pollutants are expected in both green development (SSP1-26) and the “middle” scenario (SSP2-45).²⁴ In SSP1-26, a 50% reduction in air pollutants is anticipated by mid-century, while in SSP2-45, the reduction ratio is approximately 25%. If such a substantial reduction in air pollutants is achieved, then significant changes in CH₄ lifetimes may be anticipated.

Based on the results of the sensitivity experiments, we paired the experiments and utilized linear regression to assess the relationships between the relative change in CH₄ lifetime (RCL; Equation 1) and the changes in emissions for each pollutant. The coefficients γ_x are considered to be sensitivities of lifetime to emissions (Equation 2) (For detailed information on the quantification method of sensitivity, please refer to the experimental procedures). The three sensitivity coefficients γ_x are obtained by calculating partial derivatives, which are $-0.456\%/MtN$, $0.023\%/MtC$, and $0.026\%/MtVOCs$ (Figures 1C–1E). The high R^2 of these regressions shows the robust relationship between RCL and the emissions of each pollutant (Table S2). We compare the sensitivities with the sensitivities calculated based on the data from hemispheric transport of air pollutants (HTAP) models²³ and find that they align (Table S3). The total effect of the three pollutants is calculated by summation following previous studies,^{26–28} applying a linear hypothesis (Equation 3). We validated the overall fitting formula using unused data in the fitting state and obtained an R^2 of 0.980. This indicates a robust linear fit of the formula to the chemical response of CH₄ lifetime to pollutant emissions in GEOS-Chem (Figure 1F). The robustness of this fitting linear RCL has also been verified under different CH₄ concentrations and different meteorological conditions (see Note S1 and Figure S1). This relationship is robust enough to be used in a box model as a parameter scheme for the sensitivity of CH₄ lifetime to pollutant emissions.

$$RCL = \frac{\tau - \tau_0}{\tau_0} \quad (\text{Equation 1})$$

$$\gamma_x = \frac{\partial RCL}{\partial E_x} = \frac{\partial \tau / \tau_0}{\partial E_x}, x \in [NO_x, CO, VOCs] \quad (\text{Equation 2})$$

$$RCL = \sum_{x \in [NO_x, CO, VOCs]} \gamma_x \Delta E_x \quad (\text{Equation 3})$$

CH₄ lifetime under different air quality scenarios

We applied the aforementioned parameter schemes to the OSCAR model to enhance its simulation performance in modeling the CH₄ lifetime. All subsequent experiments in the following text are also based on the OSCAR model. In this section, we analyze the differences in the CH₄ lifetime under different air quality scenarios. Here, air quality refers to the emissions of NO_x, CO, and VOC, which are ozone precursors. Aerosols are also key air pollutants, but their interaction with CH₄ is not considered in this study. We designed three air quality scenarios for discussion, labeled “clean,” “middle,” and “polluted.” In these three scenarios, CH₄ emissions are fixed to the current CH₄ emissions. The emissions of NO_x, CO, and VOCs are derived from the emissions levels of SSP1-26, SSP2-45, and SSP3-70, respectively. We used the OSCAR

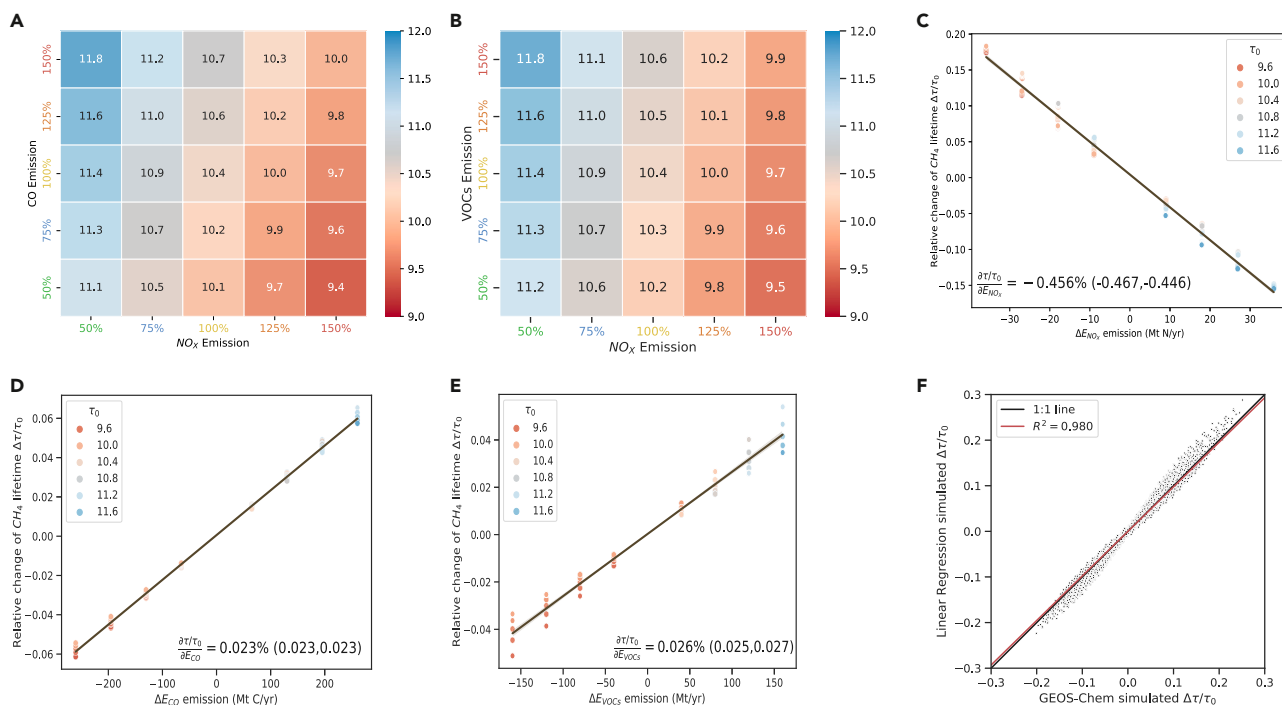


Figure 1. The sensitivity of the CH₄ lifetime to the emissions of NO_x, CO and VOCs

(A) Lifetime of CH₄ simulated by GEOS-Chem under different anthropogenic NO_x, CO emissions. Atmospheric CH₄ concentration and climate are prescribed as 2014. This sensitivity experiment is performed by varying NO_x and CO emissions by factors of 50%, 75%, 100%, 125%, and 150%, respectively, where 100% refers to 2014 emissions of 35.9 MtN/year for NO_x and 260.4 MtC/year for CO.

(B) Same as (A), but showing the combination of NO_x and VOCs, where 100% refers to 2014 emissions of 35.9 MtN/year for NO_x and 177.4 Mt/year for VOCs.

(C) The sensitivity of relative changes in CH₄ lifetime to NO_x emissions $\frac{\partial\tau/\tau_0}{\partial E_{NO_x}}$ is calculated based on the sensitivity experiment.

(D and E) Same as (C), but for CO ($\frac{\partial\tau/\tau_0}{\partial E_{CO}}$) and VOCs ($\frac{\partial\tau/\tau_0}{\partial E_{VOCs}}$).

(F) Assuming the sensitivities of the 3 pollutants to CH₄ lifetime can be linearly summed, we estimate the relative change of lifetime with Equation 3, which gives a very close result to the GEOS-Chem output.

model to simulate the RCL and lifetime under these three air quality scenarios, and their differences are attributed to variations in pollutant emissions (Figure 2).

The influence of NO_x, CO and VOCs emissions on the RCL is presented in the form of a contour map (Figure 2A). Since the sign of the influence of CO and VOCs on the CH₄ lifetime is the same and the magnitude is similar, the VOCs emissions is converted to the equivalent CO emissions (with $\gamma_{VOCs}/\gamma_{CO}$ as the factor). Positive contour lines mean that the CH₄ lifetime is extended by the pollutants, while negative contour lines mean that the CH₄ lifetime is shortened. The gray line shows the historical global anthropogenic pathway (1850–2019) from the Community Emissions Data System (CEDS) datasets.²⁹ The year 2019 is considered to be the present day and the start of the scenarios, marked as a star in Figure 2A. In comparison to the pre-industrial era (1850), the lifetime of CH₄ in the present is reduced by 7.4% due to the combined effect of increased NO_x emissions (–16.8%) and CO/VOCs emissions (9.4%). SSP1-26, SSP2-45, and SSP3-70³⁰ are considered to be the clean, middle, and polluted pathways (2020–2050), respectively. The RCL predictions for the air quality scenarios vary considerably. Compared to the present day (2019), the RCL remains almost the same under the polluted pathway, while the RCL will increase 7.0% (from –7.4% to –0.4%) under the clean

pathway (Figure 2A). Based on the linear formula, we can determine the contributions of changes in NO_x, CO, and VOC emissions and find that the change in NO_x emissions dominates the overall effect on CH₄ lifetime whether historical or future.

Even if the anthropogenic emissions of CH₄ remain as stable as 2019 (stable scenario in Table 1), changes in the emissions of NO_x, CO, and VOCs will significantly influence the CH₄ lifetime (Figure 2B). The polluted scenario has an almost constant RCL value (about –7.4%) in the coming decades in this simulation. Meanwhile, the clean scenario undergoes a rapid increase in RCL from –7.4% to –0.4% due to the reduction in NO_x emissions (Figure 2B). The difference in RCL leads to significant differences in the lifetime of CH₄; the lifetime in 2050 under the clean scenario (9.34 years) is 7.7% longer than that under the polluted scenario (8.67 years) (Figure 2B), which is a little larger than the difference in RCL (7.0%). Obviously, the emissions of ozone precursors can shape the trajectories of CH₄ atmospheric lifetime via the RCL effect. Moreover, there is a positive feedback loop here, where the longer lifetime leads to higher atmospheric concentrations of CH₄ and the higher atmospheric concentrations in turn extends the lifetime, a phenomenon known as the CH₄ self-feedback on its lifetime (see, e.g., references 1,19,23). This amplifies the RCL effect of the change in pollutant emissions. It is important to note that warming also affects CH₄

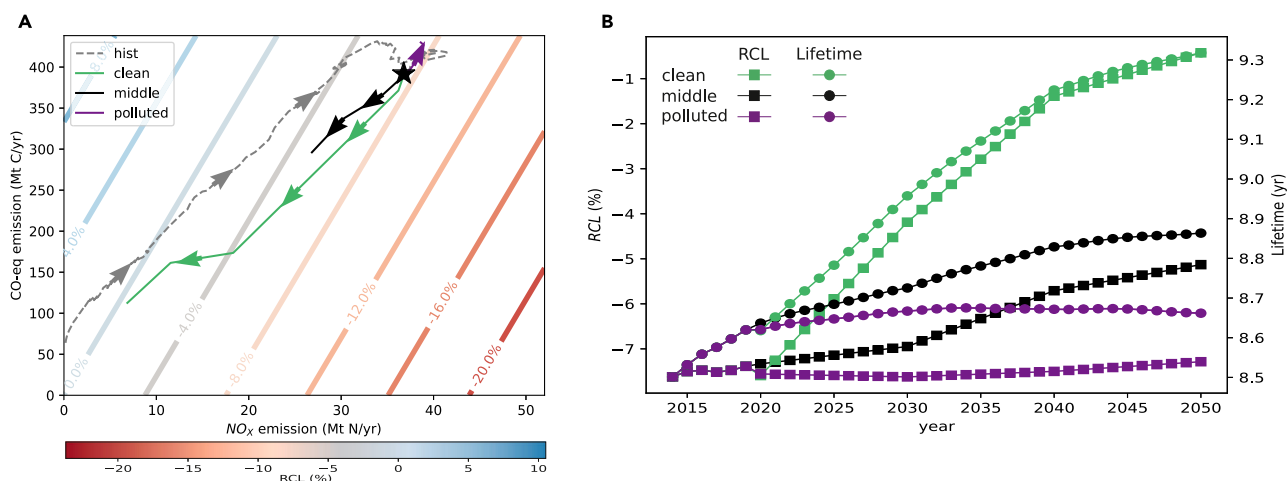


Figure 2. CH₄ lifetime under different air quality scenarios

(A) NO_x emissions shorten the lifetime of CH₄, while CO and VOCs emissions extend the lifetime of CH₄. A contour plot of the relative change in CH₄ lifetime due to NO_x and equivalent CO emissions is presented (VOCs are converted to CO here). Historical emissions and 3 future emissions pathways (clean, middle, and polluted) are plotted to show the relative change in lifetime due to the 3 pollutants (RCL). In this figure, RCL represents the relative change in lifetime compared to the pre-industrial era. The arrows above the pathways indicate the passage of time from the past to the future. The black star represents the present day (2019, the latest year of CEDS data).

(B) Trends in RCL (square markers, left axes) and CH₄ lifetimes (circle markers, right axes) for clean (green), middle (black), and polluted (purple) air quality scenarios in a fixed CH₄ emissions experiment.

lifetime. However, so as to focus on the impact of pollutants, we did not include warming effects here. Since the warming in all scenarios is similar (as the temperature change in SSP2-45 is dominated by CO₂, rather than CH₄), the changes in CH₄ lifetime due to pollutants are reliable. Other results in this study also do not account for the impact of temperature differences on CH₄ lifetime.

CH₄ reduction effectiveness under air quality scenarios

Anticipated future reductions in anthropogenic CH₄ emissions (e.g., those agreed on at the GMP) can notably and swiftly diminish atmospheric CH₄ concentrations. However, we note that an increase or reduction in the CH₄ lifetime resulting from different pollutant emissions scenarios could weaken or strengthen CH₄ mitigation efforts, respectively. Figure 3A is a conceptual diagram that qualitatively illustrates how different benefits may arise based on varying pollutant emissions for the same reduction efforts in CH₄ emissions. The purpose of Figure 3A is to demonstrate this mechanism, rather than reflect specific scenarios. In Figure 3A, the solid red line represents the trajectory of CH₄ concentration under the scenario without CH₄ emissions mitigation, serving as a reference. When mitigation efforts in CH₄ emissions are applied, the pathway of CH₄ concentration changes, such as a decrease in peak concentration and mid-century concentration and an earlier peak time. This diagram shows that under different emissions of pollutants (NO_x, CO, VOCs), the same CH₄ emissions reduction can lead to different CH₄ concentration trajectories, highlighting variations in the effectiveness of CH₄ reduction affected by pollutant emissions. In comparison to a middle pollution scenario (solid black line), in a cleaner scenario (dashed green line), CH₄ has a longer lifetime, making it more challenging to regulate atmospheric CH₄ concentration. This can be considered to be a “clean cost,”

signifying a penalty in mitigating air pollution. In other words, achieving the same reduction in CH₄ concentration in a cleaner scenario would require more effort to reduce CH₄ emissions due to the longer CH₄ lifetime. Conversely, a higher emissions scenario (dashed purple line) results in easier CH₄ control, referred to as “pollution benefit.”

The GMP is an ambitious international initiative for reducing CH₄ emissions globally.¹⁰ We have evaluated the impact of different air quality policies on the effectiveness of the GMP (Figure 3B). Based on GMP, we designed a CH₄ emissions scenario with 30% CH₄ emissions reduction by 2030 compared to 2020, with no further changes in emissions after 2030. It is combined with three air quality scenarios—clean, middle, and polluted—are considered, where the emissions of NO_x, CO, and VOCs follow the pathways of SSP1-26, SSP2-45, and SSP3-70 (see Table 1). Here, the CH₄ emissions trajectory of SSP2-45 is considered the baseline “no mitigation” scenario (red solid line in Figure 3B). In this scenario, the atmospheric concentration of CH₄ peaks at 2,114 ppb in 2044 and stabilizes at 2,103 ppb by 2050. The benefits of CH₄ emissions reduction of other scenarios are assessed by comparing against this baseline.

The other three solid lines in Figure 3B represent the GMP scenarios under different air quality policies, showing significantly lower peak and mid-century concentrations of CH₄ and earlier peak time compared to the no mitigation scenario. While they exhibit a small difference in peak concentration, a substantial disparity is observed in the mid-century concentration of CH₄. The peak concentration of the clean scenario (green solid line) is 3.1 ppb higher than that of the polluted scenario (purple solid line), both peaking 4 years after the start of CH₄ emissions reductions. The differences in peak concentration and peak time appear to be negligible. However, in 2050, the CH₄ concentration of the clean scenario is 104 ppb higher than that of the

Table 1. Scenarios defined in this study

	CH ₄ emissions ^a					Air quality ^a		
	Stable ^b	No mitigation ^c	Loose GMP ^d	GMP ^d	Strict GMP ^d	Clean ^e	Middle ^e	Polluted ^e
Clean × stable	✓					✓		
Middle × stable	✓						✓	
Polluted × stable	✓							✓
No mitigation		✓					✓	
Clean × 30%				✓		✓		
Middle × 30%				✓			✓	
Polluted × 30%				✓				✓
Clean × 20%			✓			✓		
Middle × 20%			✓				✓	
Polluted × 20%			✓					✓
Clean × 40%					✓	✓		
Middle × 40%					✓		✓	
Polluted × 40%					✓			✓

^aAll scenarios are defined as a combination of 2 parts: CH₄ emissions and air quality. Air quality in this context pertains to the emissions of NO_x, CO, and VOCs.

^bStable CH₄ emissions experiments are designed to emphasize that the change in pollutant emissions will have a significant impact on CH₄ lifetime, as illustrated in Figure 2B.

^cNo mitigation refers to CH₄ emissions in SSP2-45, which is the middle scenario and used as a reference in this study.

^dGMP stands for Global Methane Pledge. The CH₄ emissions scenario is formulated with a 30% reduction in CH₄ emissions by 2030 compared to 2020, and no additional changes in emissions are anticipated post-2030. Loose GMP and strict GMP follow a similar structure, with reduction ratios of 20% and 40%, respectively.

^eThe 3 air quality scenarios, middle, clean, and polluted, are designed with emissions of NO_x, CO, and VOCs following the pathways of SSP1-26, SSP2-45, and SSP3-70.

'polluted scenario, which must be considered. The polluted scenario has an additional reduction in CH₄ concentration of 31 ppb compared to the middle scenario (black solid line), denoted as pollution benefit. Conversely, the clean scenario bears a 74-ppb higher mid-century concentration than the middle scenario, which can be considered a clean cost. If the CH₄ emissions are increasing instead of mitigating (e.g., SSP5-85, as in Note S2 and Figure S2), then the magnitudes of pollution benefit and clean cost will be even larger (80 and 112 ppb). This underscores that changes in ozone precursor emissions will significantly impact the CH₄ pathway under different CH₄ emissions backgrounds, and the effect becomes more pronounced with higher CH₄ concentrations.

Given the uncertainty surrounding the future implementation of GMPs, CH₄ control measures may vary in stringency.

As a test, we also evaluated the impact of air pollutants on CH₄ control under more stringent conditions (40% emissions reduction, marked with triangle lines) and less stringent conditions (20% emissions reduction, marked with asterisk lines) for CH₄ emissions reductions (Figure 3B). All experiments in this section are listed in Table 1 for clarity. For a 40% reduction in CH₄ emissions, the difference in the mid-century concentration between the clean scenario and the polluted scenario is 99 ppb. For a 20% reduction in CH₄ emissions, the difference in mid-century concentration between the clean and polluted scenarios is 110 ppb. Similarly, the clean cost and pollution benefit are relatively larger for a 20% reduction in CH₄ emissions than for a more stringent reduction. This is due to the difference in atmospheric concentrations of CH₄. The higher the atmospheric concentration of CH₄, the greater the impact of the change in lifetime.

Through further simulation with CH₄ emissions reduction experiments, we find that a 35% CH₄ emissions reduction effort under the clean scenario can only achieve the similar concentration mitigation of a 30% reduction in CH₄ emissions under the middle scenario (Figure S3), showing that an additional 1/6 (~20%) effort in CH₄ emissions reduction is needed to simultaneously reduce pollution and control CH₄ due to the clean cost (Note S3).

The projection of the clean cost (the difference in the CH₄ concentration mitigation between the clean scenario and the middle scenario, where positive means less concentration mitigation) is shown in Figure 4A. The clean cost increases by about 2.4 ppb/year. By 2050, the clean cost could be up to 74 ppb. We further decompose the clean cost into NO_x reduction, CO reduction, and VOCs reduction. The overall growth trend is dominated by NO_x reduction, which would result in a cost of 152 ppb in 2050 if only NO_x abatement took place. CO and VOCs reduction have the opposite sign, as they offset costs of −39 and −37 ppb, respectively. In contrast, the projection of the pollution benefit (the difference in the CH₄ concentration mitigation between polluted scenario and middle scenario) is shown in Figure 4B. The pollution benefit grows about −1.0 ppb/year and reaches −31 ppb by 2050. The increase of NO_x emissions makes CH₄ control easier, accounting for −72 ppb. The increase in CO emissions and VOCs emissions offset 26 and 15 ppb, respectively. Overall, the change in NO_x emissions dominates the clean cost and pollution benefit, with non-negligible offset from the change in CO and VOCs emissions. There is a need to integrate the different CH₄ lifetime effects of NO_x, CO, VOCs and their climate impacts in the development of clean air policies. In addition to the aforementioned three pollutants, the emissions of new fuels

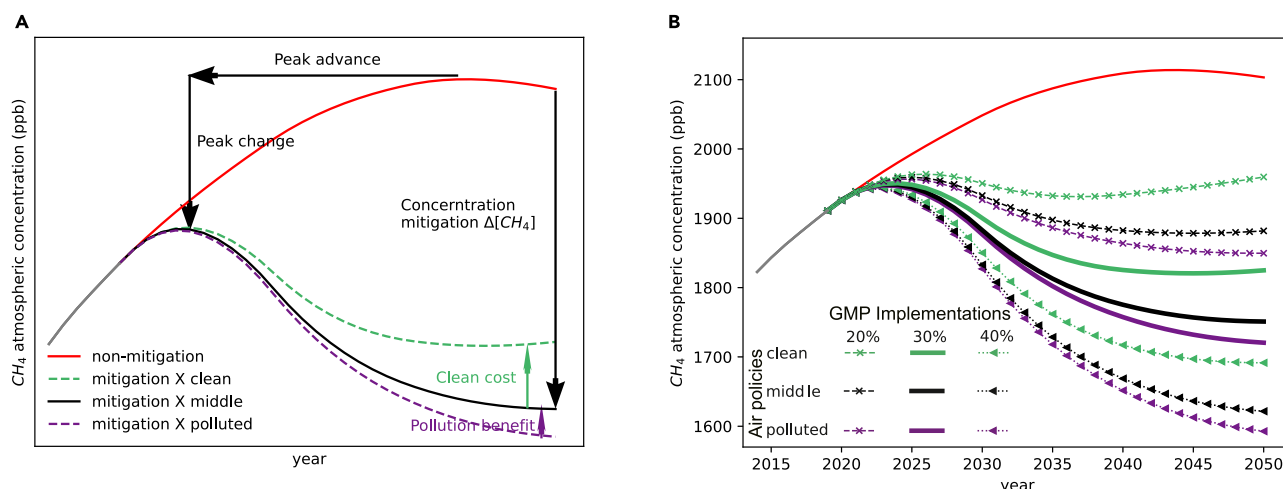


Figure 3. CH₄ reduction effectiveness under different air quality scenarios

(A) Conceptual diagram qualitatively illustrating the effect of air pollutants on the effectiveness of CH₄ mitigation. The red and black lines represent the trajectory of the CH₄ concentration under the no mitigation scenario and the mitigation scenario. The dashed green and purple lines show the effect of air quality on atmospheric CH₄ concentration control, which are referred to as clean cost and pollution benefit.

(B) The dynamics of CH₄ atmospheric concentrations for clean, middle, and polluted scenarios with different CH₄ reduction intensities. A total of 30% are the GMP CH₄ emissions reduction targets, 20% represent scenarios where the GMP is not fully met, and 40% represent scenarios where greater efforts are made to reduce CH₄ emissions than the GMP. The long lifetime of CH₄ in the clean scenario means that greater reductions in CH₄ emissions are required to achieve the same atmospheric concentrations as in the middle or polluted scenarios. The scenarios used here are defined in Table 1.

such as hydrogen³¹ and ammonia³² can also alter atmospheric oxidizing properties, potentially impacting the lifetime of CH₄. Although the current usage of these fuels is still limited, researchers may need to proactively address the effects of this atmospheric chemical mechanism.

DISCUSSION

Our findings suggest that mitigating pollutants such as NO_x has the unintended effect of increasing the lifetime of CH₄ while reducing the efficiency of CH₄ emissions reduction. This effect can significantly reduce the benefits of CH₄ mitigation, even if the GMP were fully implemented. If a comparable climate change mitigation is desired, then strengthened mitigation of CH₄ emissions will be required when pollutants such as NO_x and others are mitigated. When discussing co-benefits and trade-offs between air quality control and climate change mitigation, CH₄ mitigation is often used as an example of a co-benefit, as CH₄ is both a greenhouse gas and an ozone precursor.^{1,2} However, the effect discussed in this paper, where NO_x emissions reductions make CH₄ control more difficult, illustrates the complexity of the synergies and antagonisms between air quality improvement and climate change mitigation. To enhance air quality, it is expected that the reduction in pollutants will be the prevailing trend in the future. However, the increasing challenge of effectively controlling CH₄ due to its increased lifetime as a result of air pollution mitigation efforts will be a hurdle that humanity must confront.

Projection of future CH₄ concentrations under CH₄ emissions reduction (e.g., GMP) scenarios requires emissions-driven simulations. There are currently very few such studies. A recent study¹ based on an updated Earth system model estimated the profound, rapid, and sustained impacts of CH₄ reduction on at-

mospheric composition and climate by simulating a zero anthropogenic CH₄ emissions scenario. In our study, we calibrate the CH₄ lifetime module of a box model based on a 3D chemical transport model to quickly and accurately project future CH₄ concentrations, allowing a more comprehensive assessment of a wider range of scenarios and drivers (e.g., CH₄ reduction efforts, NO_x and other pollutant emissions).

Our results suggest that a decrease in atmospheric oxidation capacity will alter the achievable mitigation of the atmospheric CH₄ concentrations. The renewed increase in atmospheric CH₄ concentrations after 2007 or the spike in atmospheric CH₄ concentrations during the COVID-19 pandemic in 2020 have been partly attributed to a reduction in atmospheric oxidation due to changes in short-lived climate forcer emissions.^{11,14,21–23} Of particular interest are the changes in CH₄ concentrations that occurred after the pandemic, and this unintentional reduction can be seen as an experiment in voluntary emissions reduction in the future in the context of air quality control.²³

As shown in this study, the strong mitigation action scenario with low emission of air pollutants will result in a substantially longer CH₄ lifetime than the other scenarios, meaning that CH₄ concentrations will be more difficult to be mitigate. If no additional efforts are made to reduce CH₄ emissions, then this impact of mitigating pollutants on the CH₄ lifetime will inevitably reduce the benefits of the GMP. In essence, additional efforts to further reduce emissions may be needed to effectively regulate atmospheric CH₄ concentrations as intended while improving air quality.

EXPERIMENTAL PROCEDURES

Resource availability

Lead contact

Further information and requests for the model should be directed to and will be fulfilled by the lead contact, Bengang Li (libengang@pku.edu.cn).

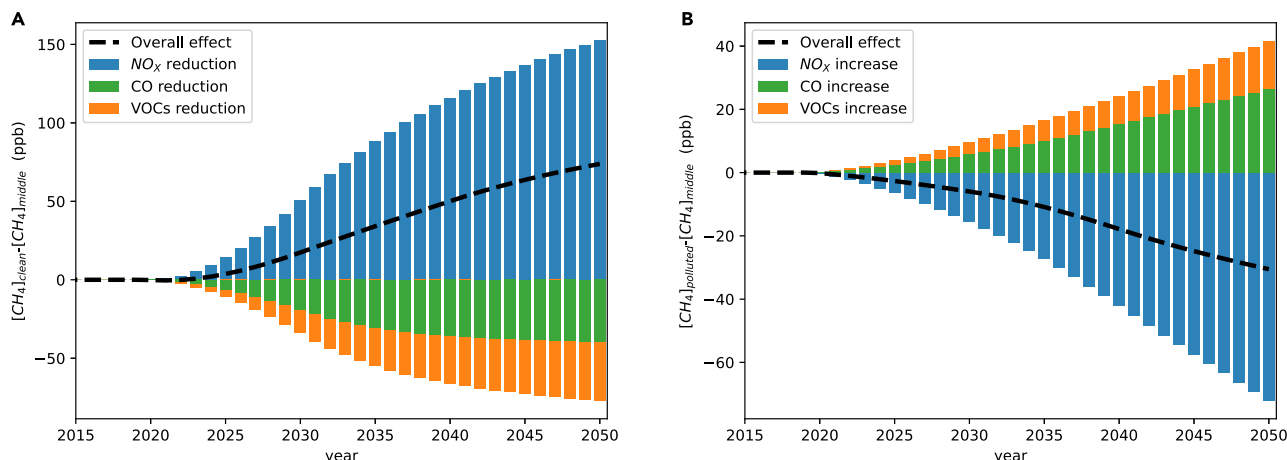


Figure 4. The decomposition of the clean cost and the pollution benefit

(A) The dashed black line shows the time series of the clean cost, which is the difference in the CH₄ mitigation between the clean scenario and the middle scenario with GMP CH₄ emissions (see Table 1). We attribute the pollution mitigation penalty to the individual ozone precursor species emissions. The clean cost is decomposed NO_x reduction, CO reduction, and VOCs reduction. The blue, green, and orange bars show the clean cost induced by only 1 pollutant reduction. (B) The same as (A), but for the pollution benefit, which is the difference in the CH₄ mitigation between the polluted scenario and the middle scenario.

Materials availability

This study did not generate new unique materials.

Data and code availability

GEOS-Chem model is available at <https://github.com/geoschem/GCClassic>

OSCAR model is available at <https://github.com/tgasser/OSCAR>

CEDS v_2021_04_21 Release Emission Data are available at <https://zenodo.org/record/4741285>

SSP Database version 2.0 is available at <https://tntcat.iiasa.ac.at/SspDb/>

All data necessary to evaluate the conclusions of this paper are listed here or in the supplemental information.

Overall

The future lifetime of CH₄ will be influenced by the anticipated reduction in pollutant (NO_x, CO, VOCs) emissions. This study aimed to assess the impact of air pollution abatement penalties on the effectiveness of CH₄ mitigation efforts. We have designed several scenarios based on SSPs and the GMP. Initially, a series of sensitivity experiments was conducted using GEOS-Chem,²⁵ a 3D chemical transport model, to simulate CH₄ lifetimes under different pollutant emissions. Subsequently, the sensitivity of CH₄ lifetime to pollutant emissions is quantified based on simulation results using regression analysis. Then, applying this sensitivity to the OSCAR model,²⁶ a box model, atmospheric CH₄ concentrations are simulated in the scenarios, and the effectiveness of CH₄ reduction under different conditions is compared.

Scenario designs

The effectiveness of CH₄ emissions reduction is the focal point of this paper, considering the influence of pollutant emissions. We designed scenarios for evaluation based on SSP²⁴ and GMP, combining scenarios of CH₄ emissions reduction with scenarios of pollutant emissions (Table 1).

All the scenarios are based on SSP2-45, with emissions of CH₄, NO_x, CO, and VOCs replaced. The scenarios of CH₄ emissions reduction include no mitigation, loose GMP, GMP, and strict GMP, reflecting different levels of enforcement of the GMP. The CH₄ emissions are set with linear mitigation over the period 2020–2030 and remain constant thereafter. No mitigation, loose GMP, GMP, and strict GMP have varying percentage reductions: 0%, 20%, 30%, and 40%, respectively. The 30% reduction aligns with the percentage stated in the GMP.

The three air quality scenarios are called middle, clean, and polluted. In the middle scenario, the emissions of NO_x, CO, and VOCs are maintained at the same levels as SSP2-45, representing a “middle of the road.” In the clean scenario, the emissions of NO_x, CO, and VOCs are substituted with those from

SSP1-26, reflecting green development. The emissions of NO_x, CO, and VOCs in the polluted scenario are replaced with those from SSP3-70, indicating a highly polluted environment.

Lifetime sensitivity quantification

The sensitivity of CH₄ lifetime to pollutant emissions is derived from the results of the GEOS-Chem model. In sensitivity experiments, NO_x, CO, and VOCs are individually designed at five emissions intensities, ranging from low to high, to cover realistic ranges for historical and SSP scenarios. Specifically, the emissions ranges are 18.0–53.9 MtN for NO_x, 130.2–390.6 MtC for CO, and 177.4–257.4 Mt for VOCs.

The emissions are based on the spatial distribution of the 2019 anthropogenic emissions but with modified global emissions. The various combinations of these emissions constituted 50 experiments (see Table S3). Using the GEOS-Chem model, CH₄ lifetimes are simulated for the 50 experiments under the same meteorological conditions (year 2014 in GCAP2 datasets), CH₄ concentrations (1,823 ppb), and scaled CEDS emissions inventories during a 1-year transient simulation. The results of these sensitivity experiments can be utilized to assess the response of CH₄ lifetime to different levels of pollutant emissions.

We hypothesize that the RCL is proportionally linked to the emissions of NO_x, CO, and VOCs, with an additive effect. This relationship is expressed as Equation 4, which is a combination of Equations 1 and 3:

$$\text{RCL} = \frac{\tau - \tau_0}{\tau_0} = \sum_{x \in [\text{NO}_x, \text{CO}, \text{VOCs}]} \gamma_x \Delta E_x \quad (\text{Equation 4})$$

In this equation, τ and τ_0 denote the CH₄ lifetime under two different experiments, and ΔE_x signifies the bias in the emissions of the respective pollutant. The sensitivities of CH₄ lifetime to the pollutant emissions (γ_x) are determined through regression analysis. The above 50 experiments are paired, resulting in a total of 1,225 possible combinations (C_{50}^2). Out of these, 200 combinations involve only 1 emission ($x = \text{NO}_x, \text{CO}, \text{or VOCs}$) differing, and they are utilized to calibrate the coefficient $\gamma_x = \frac{\partial \tau / \tau_0}{\partial E_x}$ using the least-squares method. The validation of the coefficients includes all 1,225 possible combinations, with 1,025 combinations not used in the calibration state. The overall fitting formula achieves an R^2 value of 0.98, indicating a robust linear fit of the formula to the chemical response of CH₄ lifetime to pollutant emissions in GEOS-Chem.

Effectiveness of CH₄ reduction

To simulate the time series of atmospheric CH₄ concentration and analysis of the effectiveness of CH₄ reduction, we utilize a box model called OSCAR.²⁶

OSCAR is a reduced-complexity Earth system model³³ with the capability to simulate greenhouse gas concentrations, radiative forcing, and global warming. As a reduced-complexity Earth system model, OSCAR can only simulate global mean concentration and cannot capture finer temporal and spatial distributions. The simulations in this study are conducted using a climate driven by the CO₂ concentration pathway of SSP2-45. Therefore, their climate conditions are similar.

The CH₄ module in OSCAR is determined by its emissions (anthropogenic, wetland, and biomass burning) and sinks following the mass-balance equation. The CH₄ sinks encompass four processes: tropospheric oxidation by OH, stratospheric oxidation, oxidation in dry soils, and oxidation in the oceanic boundary layer. Of these four mechanisms, that of OH oxidation is the most important. The function to calculate OH is parameterized with chemical sensitivities of OH to atmospheric CH₄, stratospheric O₃, global atmospheric temperature, and global atmospheric relative humidity. Based on the results of the above quantification, we have updated the CH₄ lifetime sensitivities to pollutants in OSCAR. Specifically, this refers to the tropospheric oxidation by the OH process, while keeping the parameters for other oxidation processes unchanged from the default settings in the original model.

Details of the CH₄ module can be found in formulas 40–42 and 47 within reference²⁶.

SUPPLEMENTAL INFORMATION

Supplemental information can be found online at <https://doi.org/10.1016/j.oneear.2024.06.010>.

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AUTHOR CONTRIBUTIONS

Conceptualization, B.L. and B.F.; methodology, software, investigation, and data curation, B.F., J.L., and Z.C.; writing – original draft, B.F.; writing – review & editing, B.F., J.L., Y.J., and B.L.; visualization, B.L. and Y.J.; supervision, B.L.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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