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The prospected air quality measurements for further unconventional natural gas developments in China based on the United States experience

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The technological innovation of horizontal drilling and high-volume hydraulic fracturing has promoted the development of unconventional natural gas (UNG) production worldwide, and hence has aroused public concern about the air pollution it may bring about. In this study, we have provided (1) an overview of the study on air pollutants from UNG emissions in the USA, focusing on both the air pollutant characterization and their related observation technologies/platforms; and (2) the potential air quality measurements of UNG development emerging in China. This study will provide useful information for Chinese environmental researchers and the local governments to deal with related air quality issues.

KEYWORDS

unconventional natural gas development, air pollution, air quality measurements, China, United States

Introduction

Nature gas is accounting for more percentage of the world's energy supply from 20% in 2000 to 23% in 2019 (https://www.iea.org), due to its relatively less environmental impact and more economic advantages compared to oil and coal (Finkel et al., 2013). To seek "energy independence," unconventional natural gas (hereafter referred to as UNG, which originates from shale, sandstone, etc.) production has been promoted extensively in the USA during recent years at the major basins containing shale gas (i.e., Marcellus Shale Play at Appalachian Basin; Fayetteville Shale Play at Arkoma Basin; Barnett Shale Play at Ft. Worth Basin, etc. Figure 1), benefiting from the significant technological innovation of the horizontal drilling and the high-volume hydraulic fracturing techniques (Finkel et al., 2013; Wang and Krupnick, 2013; Moore et al., 2014; Allshouse et al., 2017; Helmig, 2020). From 2007 to 2019, the annual UNG production increased from about 560 × 10⁸ m³ to 7900 × 10⁸ m³ with an enhancement



factor of about 14 (http://www.eia.gov/naturalgas/). However, the surge of UNG development is accompanied by a significant increase in air pollutants emissions (McKenzie et al., 2012, Shonkoff et al., 2014; Allen, 2016; Kort et al., 2016), including carbon monoxide (CO), nitrogen oxides (NOx), volatile organic compounds (VOCs, Gilman et al., 2013; Bunch et al., 2014; Helmig, 2020; Pétron, et al., 2014), primary particulate matter (PM), and their related secondary productions (i.e., ozone and secondary organic aerosols (SOA), Kemball-Cook et al., 2010; Pacsi et al., 2015; Lee et al., 2015; Liggio et al., 2016; Cheadle et al., 2017; Pozzer et al., 2020). Public concern about the air pollutants has risen with the growth of the UNG development, which has expanded into urban residential areas and has promoted related scientific studies (Pacsi et al., 2015; Adgate et al., 2014; Field et a., 2014; Vinciguerra et al., 2015; McMullin et al., 2018; McKenzie et al., 2019).

China has the most abundant shale gas resources with a total amount of 31×10^{12} m³, which is about 1.5 times of global shale gas reserves of the United States (https://www.eia.gov/analysis/studies/worldshalegas/). Although UNG production has quickly picked up in China, the annual UNG production of the USA is still about 40 times that of China in 2020 (7900×10⁸ vs 200 × 10⁸ m³ (Sun et al.,

2021). Under the most recent Chinese government energy policy, UNG production is expected to increase continually and reach 2200×10^8 m³ in 2040 (Zou et al., 2017; IEA, 2020). With the surge of UNG production, the UNG wells will inevitably expand into human communities, especially in the Sichuan Basin (Ma and Xie, 2018; He et al., 2020; Nie et al., 2020, Figure 2) which is also a population cluster with two megacities-Chongqing and Chengdu, and the subsequent air quality issues could occur. Taking the fact into consideration, we organized this study to summarize the main air pollution issues detected during the USA UNG developments and their related measurement technologies, which can provide useful guidance for Chinese environmental researchers and the local governments to better understand the air pollution-related to UNG developments.

Air pollutants from UNG production in the USA

The air pollutants caused by UNG production are mainly focused on VOCs species [including methane and Non-methane volatile organic compounds (NMVOCs)] and their related



secondary productions (including Ozone and SOA) due to their clear effect on the radiative forcing (methane) and on the human health (NMVOCs, Ozone and SOA).

Methane

Methane (CH₄) is the second-most anthropogenic greenhouse gas (after CO₂) for its effect on radiative forcing, with significant emissions from UNG operations (Helmig, 2020). Methane emissions from UNG operations were shown to be about 23 times greater than from conventional ones, due to the larger size and higher production rate of UNG (Omara et al., 2016). The reportedly estimated annual CH₄ emissions from UNG production of the main shale regions were about 158 Gg from Denver-Julesburg Basin in 2015 (Peischl et al., 2018), 270 Gg from Bakken Shale Play at Williston Basin in 2014 (Peischl et al., 2015), 131 Gg from Marcellus Shale Play at Appalachian Basin in 2013 (Peischl et al., 2015), 342 Gg from the Fayetteville Shale Play at Arkoma Basin (Peischl et al., 2015), 525 Gg from the Barnett at Ft. Worth Basin in 2013 (Karion et al., 2015), and 700 Gg from the Haynesville Shale Play at Texas-Louisiana Salt Basin (Peischl et al., 2015). The locations of each region are shown in Figure 1. Besides the CH_4 emissions from the gathering/processing plants and well pads during UNG production, CH_4 could also be emitted from the gathering pipeline, which was used to transport gases to a processing facility or a transmission line (Zimmerle et al., 2017).

Non-methane volatile organic compounds

NMVOCs from UNG production contain hazardous compounds (benzene, toluene, ethylbenzene, and xylenes (BTEX)) and other compounds (Shonkoff et al., 2014; Bolden et al., 2015), which act as precursors to O_3 and secondary organic aerosol (SOA) (Liggio et al., 2016; McDuffie et al., 2016). Enhanced NMVOCs mixing ratios near the UNG wells have been observed with variable VOCs emission rates under different

well operations (namely, drilling, fracking, coiled tubing, and flowback) (Hecobian et al., 2019). The concentration of hazardous BTEX is reported to be as high as 500 ppb downwind of oil and gas wastewater disposal facilities, which highlights the importance of monitoring BTEX concentrations during UNG operations (Helmig, 2020).

O_3 and $PM_{2.5}$

 O_3 and $PM_{2.5}$ are classified as two air pollutants on the EPA list, whose high concentration levels can cause serious harm to the human health, respiratory system in particular (Anenberg et al., 2010). The elevated level of VOCs and NOx from the UNG emissions are precursors to O_3 and SOA (Liggio et al., 2016; McDuffie et al., 2016). A number of studies have linked UNG production to nearby O_3 exceedances, even in winter (Edwards et al., 2014). The studies at the Denver basin showed about 20 ppb of O_3 enhancement (Benedict et al., 2019) and about 38% of SOA (Bahreini et al., 2018) being associated with VOCs and NOx emitted from UNG production.

Prospected air quality measurements in China

The measured air pollutants from the US shale plays based on the previous studies will be useful for narrowing the Chinese environmental researchers' focus and comparing the different emission rates of the air pollutants due to the different types of the shale basins comparing China and the United States. The measurements used in the USA also highlighted the necessity of the integration of multiple platforms for the air pollutants, including the (1) site observation platform, (2) mobile lab, drone, and airplane observation platform, and (3) satellite observation platform, for a better understanding the air pollutants emissions, while, the platforms and the related instruments mentioned below will quickly help the environmental researchers build their suitable platforms.

Site observation platform

Large-sized temporarily fixed observation sites, generally modified from mobile shelters (EPA, 2017), are most commonly used, which could support numerous monitors with a stable power supply for long-term measurements, such as (1) the commercial on-site instruments for O₃, NO/NO₂, PM_{2.5} (e.g., 2B Model 205 O₃ Analyzer, Thermo Model 42C NO/ NO₂/NOx Analyzer, GRIMM EDM 180 FEM PM_{2.5} Monitor), (2) the mass spectrometry technology-based instruments for VOCs species [e.g., IONICON Proton-transfer-reaction mass spectrometry (PTR-MS), Thermo thermal desorption gas chromatography-mass spectrometry (TD-GC-MS)] and aerosol species [e.g., Aerodyne Aerosol mass spectrometry (AMS)], (3) the laser technology-based instruments for VOCs species [e.g., Aerodyne quantum cascade lasers (QCLs)], (4) the LiDARs for O₃/PM_{2.5} vertical profiles, (5) VOCs canister collection system for further high-sensitivity VOC species analysis, and (6) the meteorological sensors (e.g. temperature, RH, wind speed, wind direction, solar radiance, precipitation). In addition to the larger temporarily fixed sites, temporary smaller sites with low-energy and low-cost sensors have also been established and widely used (https://www.epa.gov/air-sensortoolbox/evaluation-emerging-air-sensor-performance), which could compensate for the shortcomings of the larger fixed sites (e.g., lack of mobility, high cost to deploy quick-response situations or wide range). When equipped with a photoionization detector (PID) trigger system (Hecobian et al., 2019), the temporary smaller site can capture the high VOC plume using small canisters, which could provide useful information for the VOCs identification from UNG emissions.

Mobile lab, drone, and airplane observation platform

The mobile lab, generally converted from a van or a truck, can carry the above instruments to conduct on-road measurements using a multi-battery system or power generated from the engine (Boanini et al., 2021). The mobile lab could be used to capture the transport of air pollutant plumes, estimate the emission flux based on circle-route measurements, observe the spatial concentration distribution of air pollutants, act as a fixed site at some high concentration spots, etc (Mohr et al., 2011; von der Weiden-Reinmüller et al., 2014; Huang et al., 2020; Zhang J et al., 2020). However, given the terrain or the UNG factory regulations, the mobile lab could miss some areas. In this case, an unmanned aerial vehicle (UAV) carrying light-weight sensors could be used for such region observations and also for concentration vertical profile measurements (McKinney et al., 2019). The airplane platform could be used for large-scale observation, but the number of flights or aircraft could be limited due to the high cost.

Satellite observation platform

Satellite observations could provide temporary and spatial concentration variation of some species, such as NOx, CH_4 , O_3 , etc, which could be used to evaluate the influence of UNG emissions on a large scale (Jacob et al., 2016; Goldberg et al., 2019; Varon et al., 2019). Combining the model simulation using existing emission inventories and comparing it with the current satellite observations, the current species emission rate could be estimated (Zhang Y et al., 2020), which would be very useful for updating the existing emission inventories.

Conclusion

Benefiting from the horizontal drilling and hydraulic fracturing techniques innovation, the USA has experienced the boom of the UNG development and reached its goal of "energy independence." However, UNG development in China is still in its infancy and has a great potential for rapid growth, which will inevitably result in air pollution, as has been seen in the Denver Basin. These issues should be given top priority due to the clean air action from the current Chinese government. The studies on the air quality in the USA for the UNG-related air pollutant emissions provide solid knowledge, either on the air pollutants characterization or their related observation technologies/ platforms, which can provide very useful guidance for Chinese environmental researchers and the local governments to deal with the air quality issues caused by the further surging of UNG production.

Author contributions

Conceptualization, formal analysis, TF and JZ; investigation, resources, TF, XZ, SQ, and JF; writing—original draft preparation, TF; writing—review and editing, XZ, SQ, and JZ. All authors have read and agreed to the published version of the manuscript.

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Conflict of interest

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