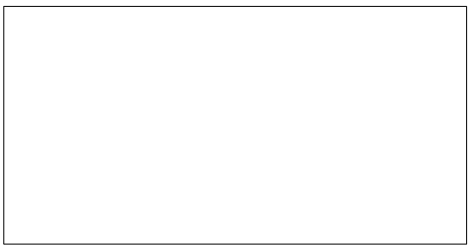

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Cryogenic circulation for indoor air pollution control

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Abstract: Hazardous outdoor air pollution has severely affected indoor air quality, threatening the health of billions of people. However, existing indoor air purification technologies are unsatisfactory due to some inherent limitations such as poor efficiency, limited target pollutants, the need to frequently replace filters or adsorbents, or the generation of harmful by-products. Here, we studied the effect and mechanism of cryogenic circulation for indoor air purification. Experimental results show that up to 99% of indoor $PM_{2.5}$ from ambient air was removed at -18℃. The morphological measurements indicate that micrometer-sized particles are formed concomitantly with the reduction of nanometer- or submicron-sized particles, suggesting that condensational growth of fine particles is responsible for the removal. Applying the method to gaseous pollutant purification demonstrates that 98% of NO_2 is condensed and removed from the ambient air at -50℃, implying that the method would be effective for multiple indoor pollutants with higher boiling points. Cryogenic condensation may provide a principle for continuous indoor air purification via modified air conditioners and humidifiers in cases where health benefits outweigh energy consumption concerns.

Keywords: $PM_{2.5}$ pollution; Air cleaner; Public health; Indoor air quality

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1. Introduction

Severe haze pollution has become a "global plague" (Huang et al., 2014; Wang et al., 2014). Chronic exposure to haze pollution contributes to weight gain, cardiorespiratory and metabolic dysfunction, respiratory problems and impaired mental health (Lim et al., 2012; Shin, 2016; Wei et al., 2016), leading to 3.3 million premature deaths annually worldwide (Jerrett, 2015; Lelieveld et al., 2015). Hazardous outdoor air pollution in many countries under rapid industrialization has severely affected indoor air quality (Kulmala, 2015; Luengas et al., 2015). Many people spend more than 90% of their time indoors, particularly if outdoor air quality is poor (Kulmala, 2015), therefore indoor air quality in regions with haze problems is a vital environmental factor that affects public health.

The present state of indoor air purification is unsatisfactory due to various limitations of the existing technologies (Luengas et al., 2015). Mechanical and electronic filtration are effective for removing suspended particles from indoor air, but frequent replacement of filters is required to maintain good performance (Luengas et al., 2015). Adsorption has been applied to retain gaseous contaminants, but the regeneration of adsorbents involves significant additional costs (Jo and Yang, 2009; Luengas et al., 2015). Photo-oxidation promotes the degradation of indoor organic contaminants, but the method is often pollutant-specific and short lived, and may generate harmful by-products (Destaillats et al., 2012; Ourrad et al., 2015). Therefore, there is a need for sustainable methods that can continuously remove air pollutants at the cost of reasonable energy consumption.

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Natural "wet deposition" (i.e., precipitation reaches the Earth's surface), is a major removal pathway of aerosol particles from the atmosphere (Pöschl, 2005). Precipitation originates from cloud particles, which are formed by condensation of water vapor on aerosol particles (Baker and Peter, 2008; Hoose and Mohler, 2012; Pöschl, 2005). Inspired by this phenomenon, we hypothesize that phase transition of gaseous components by cryogenic methods may be used to separate the haze particulates and gaseous pollutants from polluted air.

Although cryogenic methods have been applied to distill or concentrate industrial volatile organic compounds, in most cases the flue gases are at high concentration (>1%) (Dwivedi et al., 2004; Gupta and Verma, 2002), and hence the situation is different from the indoor environment with both gaseous and particulate pollutants at trace levels (Luengas et al., 2015; Wainman et al., 2001). Although refrigeration equipment is widely available worldwide, we found no specific studies on the use of cryogenic methods to tackle indoor air pollution. Here, we studied the effect and mechanism of cryogenic circulation in removing both indoor airborne particulate matter (PM) and typical associated gaseous pollutants NO_2, and analyzed the technical challenge for practical applications.

2. Materials and methods

2.1 Experimental set-up

The polluted air was subjected to a cryogenic process when introduced into a single-tube circulated stainless steel pipeline installed in a custom-designed box

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condenser (50 cm×50 cm×70 cm), as shown in Fig. 1. The condenser was made up of a liquid nitrogen injection module, an intelligent temperature control module and a fan. Gas flow of liquid nitrogen vapor was controlled by the intelligent temperature control module to maintain the inner of the condenser at a given temperature (-60–25℃). The diameter (1.6 cm) and length (15 m) of the pipeline were designed to prolong the reaction time of the polluted air in the pipeline. Laminar flow conditions of the inlet polluted air were ensured during the experiments. The pipeline was restored to normal temperature and washed by clean air before each cryogenic treatment.

2.2 Indoor air fine particle removal

Polluted indoor air in the Chinese laboratory was used for the study during heavy outdoor haze events between December 1, 2014 and January 28, 2015. The indoor temperature and relative humidity were 20–23℃ and 20–30% during the period (central heating applied in the laboratory), respectively. The indoor real-time haze samples were divided into three pollution levels according to the $PM_{2.5}$ (particles of $d \leq 2.5$ μm): extremely heavily polluted (EHP, $PM_{2.5} > 0.50$ mg/m^3), heavily polluted (HP, 0.50 mg/m^3 > $PM_{2.5}$ > 0.15 mg/m^3) and lightly polluted (LP, 0.15 mg/m^3 > $PM_{2.5}$ > 0.075 mg/m^3).

In the blank experiments, haze polluted air with different pollution levels passed the condenser pipeline without cryogenic treatment at a volume flow rate of 2 L/min, to estimate the scavenging phenomena of particles occurring within the pipeline when

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a temperature gradient is absent. The indoor real-time haze polluted air was then introduced into the condenser at a designed temperature and subjected to a cryogenic process at the same volume flow rate. During the experiment, thermocouples and hygrometers (Testo 635-2, Germany) were installed to monitor the temperature and humidity of gas flow at the inlet and exit of the condenser. Concentration and size of aerosols at the inlet and exit were continuously and simultaneously monitored, and the removal efficiency for a single-pass was calculated accordingly. Moreover, haze samples before and after cryogenic treatment were directly collected on conductive black tape for TEM testing, to examine the morphological variation.

The mass concentrations of PM_{10} (particles of $d \leq 10$ μm), $PM_{2.5}$ and PM_1 (particles of $d \leq 1$ μm) were measured using an LD-5C microcomputer laser dust monitor (Greenwood Innovation Co., Ltd, China), with a detection range of 0.001–10 mg/m^3. The particle number concentrations of 0.30–10 μm were detected by a CI-208 particle analyzer (Climet Co., USA). The airflow in the exit was thawed by heater bands with a temperature controller before detecting the mass and number concentration of the aerosol particles.

2.3 NO_2 removal

Highly purity N_2 was used to dilute the NO_2 to desired initial concentrations of 0.42, 0.66, 1.41, 5.78 and 9.23 ppm. The NO_2 gas was passed through the condenser at a gas flow velocity of 2 L/min under different cryogenic temperatures. The NO_2 concentrations of the gas mixture were detected on the inlet and exit of the condenser,

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and the single-pass efficiency of NO_2 was obtained by calculating the NO_2 concentration changes during a cryogenic process at a given temperature. The concentrations of NO_2 were monitored by a Digital Compact Portable NO_2 Analyzer (Interscan Corporation, USA).

3. Results and discussion

3.1 Removal of haze particles

When the haze-polluted air was introduced into the condenser, the mass concentration of haze particles at the exit decreased with decreasing cryogenic temperature (Fig. 2d-f) compared to that of PM_1, $PM_{2.5}$ and PM_{10} at the inlet (Fig. 2a-c). In the blank experiments (20℃), 20-30% of micron size particles PM_{10} and $PM_{2.5}$ were removed by gravitational settling within the pipeline (Nishio et al., 1974). Intriguingly, the mass concentrations of PM_{10} and $PM_{2.5}$ were reduced to below the Chinese national standard levels (0.15 mg/m^3 for PM_{10} and 0.075 mg/m^3 for $PM_{2.5}$) at -15℃ and -16.5℃, respectively. PM_1 particles are suspected to pose the greatest health risk among the respirable particles (Sun et al., 2014; Tian et al., 2014). Due to the dominant deposition effect of submicron particles — Brownian diffusion was negligible (Fuchs, 1964; Batchelor and Shen, 1985), PM_1 particles were found hardly removed within pipeline in blank experiments. However, half of the PM_1 particles were removed from LP samples and nearly all of them were removed from EHP samples at temperatures lower than -18℃. These results indicated that, cryogenic circulation was effective in removing both micron and submicron size particles from

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indoor polluted air, and the removal efficiency of fine particles is markedly higher at higher pollution levels.

The number concentration of haze particles was detected (see Supplementary Fig. S1) and particle number evolution of size-resolved particles during the cryogenic treatment is illustrated in Fig. 3a. Taking EHP samples as an example, when the haze samples passed through the condenser pipeline but without cryogenic treatment (at 20℃), less than 10% of fine particles of d = 0.3–1 μm were removed. When the haze-polluted air was cooled to around -10℃, distinct reductions in the concentrations of particles of d = 0.3–0.5, 0.5–1 and 1–3 μm were accompanied by successive increases in coarse particles of d = 5–10 and >10 μm. These results imply that coarser particles were formed by coagulation of the multiple fine particles. This result was clearly confirmed by the morphological variation of haze particles during the cryogenic process around -10℃ (Fig. 3b and 3c), during which a considerable number of nanometer- or submicron-sized particles were found to be replaced by particles of several microns in diameter. Additionally, water moisture in the haze flow was decreased to zero around -10℃ (Fig. 3d), suggesting water vapor was effectively condensed under cryogenic treatment.

3.2 Removal of NO_2

In addition to particulate pollutants, the gaseous pollutant NO_2 at relatively low concentrations (ppb to ppm levels) was treated at various cryogenic temperatures. NO_2 was the focus because it has been shown to be a ubiquitous indoor pollutant

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(Challoner and Gill, 2014; Cibella et al., 2015; Xie et al., 2015), and high levels of NO_2 are associated with increased respiratory symptoms and reduced lung function (Cibella et al., 2015; Kattan et al., 2007). Fig. 4 shows that the NO_2 concentration decreased remarkably with decreasing temperature, and -50℃ was sufficient to meet the Chinese indoor standard of 0.12 ppm under different initial NO_2 concentrations (0.42–9.23 ppm). In addition, the removal efficiency for NO_2 was significantly enhanced at higher pollution levels, due to correspondingly higher mass transfer rates from the bulk vapor phase to the liquid or solid condensate (Dwivedi et al., 2004).

3.3 Purification mechanism

When the polluted haze air passed through the condenser, thermophoresis and cryogenic condensation are the two possible explanations for the enhanced particle removal inside the cooled pipelines. Thermophoresis is a common phenomenon whereby aerosol particles drift towards and deposit onto the wall of the pipeline when subjected to a temperature gradient (Lin et al., 2008). It is found particularly effective in removing submicron particles from gas and their removal efficiency depends on the temperature gradient (Batchelor and Shen, 1985; Wang and You, 2013). Usually, less than 10% of the inlet dried submicron particles are deposited by thermophoresis when they pass though pipelines with temperature gradient of 30-80℃ under laminar flow condition (Montassier et al., 1991; Nishio et al., 1974; Romay et al., 1998; Stratmann et al., 1994; Tsai et al., 2004; Wang and You, 2013). This value was much lower than the removal efficiency of PM_1 in our experiment, for example, nearly all PM_1 were

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removed from EHP samples when the temperature gradient was 38℃ (from 20℃ to -18℃) (Fig. 2). Therefore, thermophoresis deposition was not the principal removal mechanism of haze particles by this cryogenic method.

It is known that water vapor in homogeneous systems can become ice at temperature as low as -38 ℃, however, this process can be triggered by aerosol particles acting as ice nuclei at much higher temperatures below 0℃ (Pruppacher and Klett, 2012). The latter process is defined as heterogeneous condensation of water vapor, and it is a useful pre-conditioning technique for fine particle enlargement (Heidenreich and Ebert, 1995; Yang et al., 2010). In this study, heterogeneous water nucleation on the fine haze particles was achieved by cryogenic treatment, and distinct formation of micron-sized particles of d = 1-3, 3-5 and 5–10 μm began at around -10℃ (Fig. 3 and Supplementary Fig. S1). This temperature coincided with the onset freezing temperature of some typical aerosols, for example, bioaerosols, soil particles and clay mineral coated with soluble ions (Fornea et al., 2009; Hoose and Mohler, 2012). Therefore, the condensational growth of PM_1 was responsible for the reduction of PM_1 exit mass concentrations compared to $PM_{2.5}$ and PM_{10} at -10℃ (Fig. 2). Moreover, fine particle enlargement is significantly enhanced by increasing the temperature gradient between the inlet gas and cooled pipeline (Tammaro et al., 2012), suggesting larger micron-sized particles (eg. particles of d >10 μm) were formed by cryogenic growth and collision/agglomeration among particles (Fig. 3 and Supplementary Fig. S1). The coarser particles are prone to be separated from the polluted air and removed by gravitational or inertial settling within the pipe (Tammaro

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et al., 2012; Yang et al., 2010). Therefore, the remained fine particles PM_1 become dominant in the exit air (Supplementary Fig. S1), and the corresponding exit mass concentration of nearly all the particle matter fractions were more or less the same at lower cryogenic temperatures (Fig. 2). Overall, heterogeneous condensation was mainly responsible for the removal of fine particulate pollutants under cryogenic treatment.

For purely gaseous pollutant NO_2 with low concentration, cryogenic treatment was also a feasible removal method (Fig.4). However, a temperature lower than boiling points was needed to achieve the homogeneous condensation condition (Dwivedi et al., 2004). In the real indoor environment, the composition of polluted air is much more complex, including particles of different size and composition, inorganic gaseous compounds (eg., NO_X) and organic gaseous compounds (eg., VOCs), etc. Many indoor organic compounds (e.g., toluene, benzene, xylenes) have much higher boiling points (50–260℃) than NO_2 (21℃) (Dwivedi et al., 2004; Luengas et al., 2015). Therefore, they can be condensed and removed by cryogenic condensation at higher cryogenic temperatures than NO_2.

3.4 Technical consideration for practical application

As a proof of concept, we studied the purification capability and mechanism of cryogenic principle for indoor air purification. This may evoke the development of new generations of air conditioners that can not only control the temperature but also the quality of indoor air (Pan, 2017). However, use of the cryogenic principle will

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have to involve energy consumption. In order to assess the energy cost, we used the recommended clean air delivery rate (CADR) of 200 m^3/h (Shaughnessy and Sextro, 2006) to estimate the energy consumption of the cryogenic circulation method (see Supplementary Information). It showed that a minimum energy consumption of 2.59 kW was required to cool the polluted air (with humidity of 20%) from 20 to -18 ℃. This energy consumption value was about 12–65 times higher than that of high-efficiency particle arresting (HEPA) air cleaning, 25 times higher than that of electrostatic precipitators (ESP), and 6–16 times higher than that of ventilation with filter systems (Table 1). The high energy consumption of our cryogenic principle may be reduced in practical application if more energy-efficient refrigerants or materials are developed (Azmi et al., 2017; Koizumi, 2007; Matsumoto and Omata, 2017). Before it can be considered for domestic applications where substantial energy reduction is required, cryonic circulation can be used for continuous indoor air cleaning when health is prioritized above energy consumption, such as in nuclear submarine or industrial underground operations.

Additional operational challenges need to be further studied before the cryogenic principal can be used for practical purposes. For example, pipeline congestion due to the condensed ice particles, power source for reheating the cooled air to room temperature, or how to maintain comfortable indoor humidity during cryogenic processes. Commercialization of the cryogenic method for air purification represents both a business opportunity and a production challenge.

4. Conclusion

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This study demonstrated the effectiveness of cryogenic circulation for removing both particulate and gaseous pollutants from indoor air, and cryogenic condensation was the principal removal mechanism. However, energy consumption, humidity control and other technical challenges should be further studied before practical application.

Acknowledgement

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Figure legends:

Fig. 1. Schematic diagram of experimental facility

Fig. 2. Removal efficiency of haze particles at LP, HP and EHP pollution levels in ambient air. (a-c) Inlet mass concentrations of PM_1, $PM_{2.5}$ and PM_{10}; (d-f) Exit mass concentration of PM_1, $PM_{2.5}$ and PM_{10} as a function of cryogenic temperatures. The red straight lines indicate the Chinese ambient air quality standards for daily average levels of PM_{10} (0.15 mg/m^3) and $PM_{2.5}$ (0.075 mg/m^3).

Fig. 3. Evolution of haze-polluted air at EHP level during the cryogenic treatment. a) Variation of particle number concentrations for size-resolved particles under different cryogenic temperatures. b) TEM images of haze particles before cryogenic treatment; c) TEM images of haze particles after cryogenic treatment at around -10℃; d) Variation of specific humidity under different cryogenic temperatures.

Fig. 4. NO_2 exit concentration as a function of cryogenic temperature. The red straight line indicates the Chinese indoor standard for instantaneous NO_2 concentration.

Table legends:

Table 1 Features of existing air cleaning methods.

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Table 1 Features of existing air cleaning methods.

Air cleaning method	CADR (m^3/h)	Electrical power consumption (kW)	Data source
HEPA[a] air cleaning	180-324	0.04-0.206	(Noh and Yook, 2016; Peck et al., 2016; Waring et al., 2008)
Air ionic cleaning	200	0.032[c]	(Siegel, 2016; Waring et al., 2008)
ESP[b]	284	0.102	(Waring et al., 2008)
Ventilation/filter system	360	0.164-0.425	(Noh and Yook, 2016)

[a]: High-efficiency particle arresting; [b]: Electrostatic precipitator; [c]: Estimated through operation cost effectiveness of ion generator (Siegel, 2016), by assuming CADR to 200 m^3/h.

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Highlights

- Cryogenic condensation can remove indoor fine particulate and gaseous pollutants.
- Phase transition of gaseous components induces haze pollutants removal.
- Obtain clean indoor air for health through reasonable energy consumption.

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