



Quantifying Influences of Nocturnal Mixing on Air Quality Using an Atmospheric Radon Measurement Case Study in the City of Jinhua, China

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ABSTRACT

The atmospheric mixing state and emission rates play decisive roles in public exposure to urban air pollution. This study utilizes atmospheric radon measurements taken with the SM200 “stability monitor,” which reflect changes in the atmospheric mixing state, to evaluate and forecast air quality. Using six months (March–August 2016) of atmospheric radon measurements in Jinhua, China, we classify the nocturnal atmospheric stability conditions into four distinct categories, “well-mixed”, “weakly stable”, “moderately stable”, and “most stable”, by applying a modified radon-based stability technique. We calculate the atmospheric self-cleaning ability index (ASI) and evaluate it with the four-category stability scheme, and the results confirm that the atmospheric radon measurements reliably represent the atmospheric mixing state. Analyzing PM_{2.5}, PM₁₀, SO₂, NO₂, CO, and O₃ measurements from three nearby stations during the campaign, we find that the pollutant concentrations and air quality index (AQI) values assigned using the aforementioned stability scheme are consistent with the defined atmospheric mixing states. We subsequently demonstrate that the modified radon-based stability method is suitable for targeting the most unfavorable air quality conditions and determining where the emissions originated. Finally, we propose a simple ASI-based model for predicting regional severe air pollution.

Keywords: Nocturnal mixing state; Atmospheric radon measurement; Atmospheric self-cleaning ability; Air quality prediction.

INTRODUCTION

Atmospheric fine particulate matter (PM_{2.5}) is the most important air pollutant in China, contributing to a premature mortality rate of ~1 million year⁻¹ (Lelieveld *et al.*, 2013). The entire population of China has been exposed to PM_{2.5} concentrations above the World Health Organization (WHO) annual mean guideline value of 10 mg m⁻³. And the population lived in areas that met the annual PM_{2.5}, PM₁₀, NO₂, and SO₂ National Ambient Air Quality Standards (NAAQS) Grade II (Song *et al.*, 2017) are 7% (14%, 19%), 17% (27%, 34%), 51% (67%, 70%), and 88% (97%, 98%) in 2014 (2015, 2016), respectively.

The concentrations of most air pollutants have been decreasing since 2013 and as a result southern China has had less air pollution (Zhao *et al.*, 2018a, b; Wu *et al.*, 2019b). Efforts to reduce emissions have shown benefits in many places in China since a series of air quality control measures were implemented during the 11th and 12th Five Year Plan (2006–2010 and 2011–2015), and specific actions during some periods like the Olympics, APEC, etc. (e.g., Wang and Hao, 2012; Zhang *et al.*, 2016; Wu *et al.*, 2017; Wang *et al.*, 2019a). In 2013, the central government launched the Clean Air Act (CAA) and identified binding reduction targets for emissions of SO₂ and NO_x for each city in order to mitigate PM_{2.5} pollution in China (MEPC, 2015). However, air pollution in China is still a serious problem, and 70.7% of the 338 cities in China did not meet the national standard in 2017 (MEE, 2018). Although Beijing reached the target value of 60 µg m⁻³ for PM_{2.5} in 2017, a large gap still remains between this target and the criteria of the WMO (10 µg m⁻³ for annual means; 35 µg m⁻³ for daily means).

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There are huge challenges for China to reach the national air quality standards, since: (i) the overall emissions are still very large, (ii) emission sources are still very complex, and (iii) understanding of the underlying processes for haze formation and identifying effective mitigation pathways is still poor (Horst *et al.*, 2018; Wen *et al.*, 2018; Wu *et al.*, 2019a; Yang *et al.*, 2019). Meteorological conditions are the primary contributing factor to day-to-day variations in pollutant concentrations. Air quality is very sensitive to unfavorable meteorological conditions in many cities in China and severe haze episodes occur once there is stagnant or stable air (He *et al.*, 2016; He *et al.*, 2017; You *et al.*, 2018; Zhao *et al.*, 2018b; Mei *et al.*, 2019). It is important to quantify meteorological influences on air quality in order to better evaluate emission reduction measures (e.g., Chambers *et al.*, 2019a). It is also important to predict pollution episodes in a relevant longer time (like two weeks or one month before) in order to enable regional air pollution prevention actions (Zhang *et al.*, 2017; Wang *et al.*, 2019b). Quantitative descriptions of the atmospheric pollution dispersion equation have been developed for air pollution forecasting. Xu and Zhu (1989, 2000) defined the atmosphere ventilation equation and developed a forecast model for short-term meteorological conditions of air pollution. The ventilation coefficient was also used to estimate air pollution potential (Gassmann and Mazzeo, 2000; Ashrafi *et al.*, 2009). Wang *et al.* (2012) established the PLAM (Parameters Linking Air quality to Meteorological elements) index to evaluate the meteorological influence on PM₁₀ concentration in Beijing from 2000 to 2007. Zhu *et al.* (2018) developed the atmospheric self-cleaning ability index (ASI) and used it for long-term forecast. A common feature of all indices for assessing the long-term and short-term emission reduction measures is trying to synthetically assess the atmospheric mixing state based on numerous measurements of individual meteorological parameters. These indices are modeled and introduce uncertainty in their calculation process. Atmospheric radon measurements can provide an index of the atmospheric mixing state, which has been used for air pollution assessment (e.g., Perrino *et al.*, 2001, 2012; Chambers *et al.*, 2015b; Wang *et al.*, 2016). Radon-222 (radon) is a unreactive, poorly soluble gas and atmospheric radon measurements have been used as an indicator of atmospheric stability and mixing for over one hundred years (e.g., Wigand and Wenk, 1928; Moses *et al.*, 1960). Recent studies have addressed the observations of radon concentration vertical gradients (e.g., Chambers *et al.*, 2011; Williams *et al.*, 2011), interpretation of temporal changes in radon concentration (e.g., Singh *et al.*, 2005; Podstawczynska, 2016), the estimation and removal of fetch effects (e.g., Chambers *et al.*, 2015; Williams *et al.*, 2016), and established radon-based stability index as directly representative of the combined near-surface physical atmospheric mixing processes (Williams *et al.*, 2013; Chambers *et al.*, 2015; Wang *et al.*, 2016).

The combined use of modeled ASI values and observed radon-based stability classification can help to better understand the atmospheric mixing state, its influence on air quality, and how it may be used for pollution mitigation purposes. This paper aims to (i) evaluate the ASI index using

radon-based stability classification, (ii) quantify the nocturnal mixing state influence on air pollution using an improved radon-based atmospheric stability technique, and (iii) predict air quality using radon measurements and ASI.

EXPERIMENT

Site and Instrumentation

Atmospheric radon was measured in the city of Jinhua, China, from March 2016 to August 2016. Air quality concentrations of the six criteria pollutants (PM_{2.5}, PM₁₀, SO₂, NO₂, CO and O₃) were monitored at three stations, A, B and C, of the national air quality monitoring network in Jinhua city, and the data used in this study are from the China Air Quality Online Monitoring and Analysis Platform (<http://datacenter.mep.gov.cn>, <http://106.37.208.233:20035>). The location of the stations is shown in Fig. 1.

- Station A (119.649°E, 29.113°N), representative of traffic areas, northeast to Wuyi Road;
- Station B (119.647°E, 29.077°N), representative of urban residential;
- Station C (119.686°E, 29.103°N), representative of urban residential, background regions;
- Station D (119.806°E, 29.166°N), northeast to Jinhua city center.

CMA (China Meteorological Administration) (119.65°E, 29.1167°N), near to Station A, is the regional background station operated by China Meteorological Administration.

Natural radioactivity of radon has been conducted above ground level (a.g.l.) at Station D (Fig. 1) using an SM200 “stability monitor” (OPSIS AB, Furulund, Sweden). The instrument consists of a particulate matter sampler with two moving sampling filters and beta measurement by a Geiger-Muller counter for determining the total beta activity of the short-lived radon progeny. This instrumental feature assures that the short-lived beta activity of the particles is determined continuously over an integration time of 4 hours and that the beta measurement period is long enough to guarantee a good accuracy of the results (Perrino *et al.*, 2001). The output from the SM200 is raw counts, not a calibrated concentration, and will henceforth be denoted *Rn* to indicate that it is a proxy only of ambient radon progeny activity (refer to “radon” in the following text). Climatological observations (wind speed, wind direction, relative humidity (%), temperature and atmospheric pressure) were made by the CMA at the site named CMA in Fig. 1. In this study, hourly values of pollutants (NO₂, CO, SO₂, PM_{2.5}, PM₁₀), and meteorological data are analyzed.

Atmospheric Radon and the Stability Classification Scheme

The SM200 monitor in Jinhua recorded 4-hour *Rn* measurements during this campaign and the data is used for atmospheric mixing status analysis. The stability classification scheme using SM200 monitor was reported in our previous studies (Chambers *et al.*, 2015; Wang *et al.*, 2016). We first approximated and removed fetch effects (see details in Chambers *et al.*, 2016), then sort the daily variation for stability classification. The radon-based stability classification

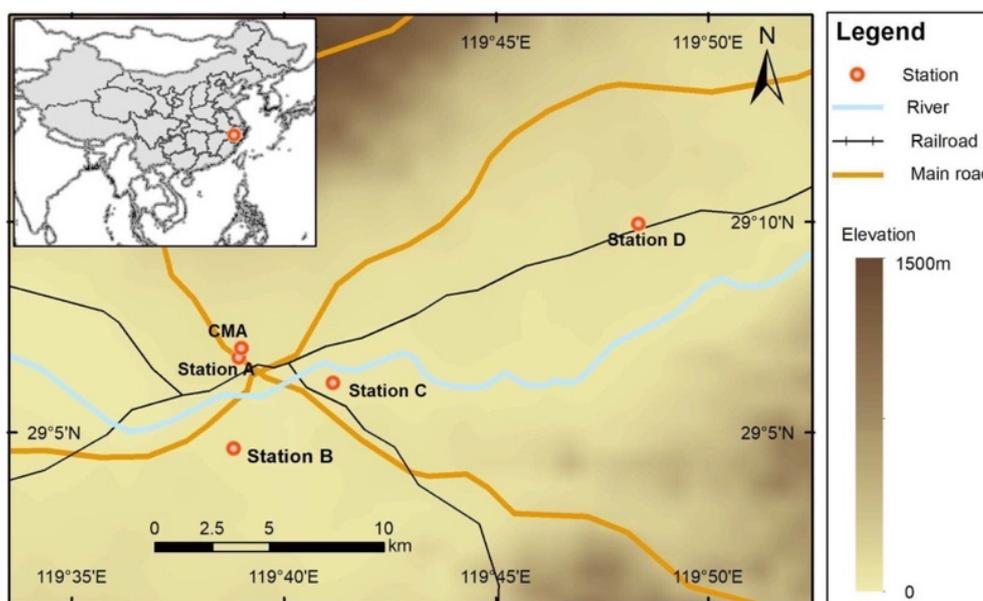


Fig. 1. Location of meteorological and pollution measurement sites.

technique uses nocturnal mean radon data each day within an 8-hour nocturnal window from 20:00 to 04:00 (2000–0400 h; Fig. 2(a)) as a proxy for the combined influences of all near-surface nocturnal atmospheric mixing processes. The nocturnal mean radon concentrations (Rn^*) are referenced to the corresponding 2000h data, in order to minimize seasonal effects on the absolute magnitude in Eq. (1).

$$Rn_{2000-0400h}^* = \frac{\sum_{i=1}^3 Rn_i}{3} - Rn_{2000h} \quad (1)$$

According to the cumulative frequency of nightly mean Rn^* values (Fig. 2(b)), four nocturnal mixing categories (or “stability”) are defined: well-mixed moderate to strong winds, often overcast, rainfall is common; weakly stable light to moderate winds, cloudy or overcast, occasional rainfall; moderately stable light winds, some scattered cloud, mostly clear, rainfall very uncommon; and most stable calm to light gradient wind, clear skies, no rain. Negative Rn^* values associated with passing strong synoptic systems or fronts are not included. Similar radon-based stability classification using the AlphaGUARD detector has been evaluated by Chambers *et al.* (2019b) in the classification of the urban surface layer.

Atmospheric Self-cleaning Ability Index (ASI)

The ASI was defined based on the prediction principle of the City Air Pollution Prediction System (CAPPS), which is an index to quantitatively describe air pollution meteorological conditions using Eq. (2) (Xu and Zhu, 1989; Zhu *et al.*, 2018). The ASI represents physical self-purification of the atmosphere, which reflects the atmospheric environmental carrying capacity of emissions, and is used as reference to control regional emissions and project air pollution potential (Han *et al.*, 2017; Xu *et al.*, 2017):

$$ASI = Q/S = \left(\frac{\sqrt{n}}{2} V_E + W_r R \sqrt{S} \right) \cdot C_s / \sqrt{S} \quad (2)$$

where Q is air volume of a pollutant; S is unit area; V_E is ventilation based on wind and boundary layer height; C_s is the standard concentration of air pollutant; W_r is washout constant (6×10^5) and R is precipitation.

The air pollution prediction equation used in CAPPS (Eq. (3)) takes into account hourly meteorological parameters and the accumulating effect of air pollution. The parameters including wind, mixing height and precipitation used to calculate ASI are outputs from the Weather Research and Forecasting (WRF) model, detailed description in Zhu *et al.* (2018):

$$ASI = ASI_t \left(1 - e^{-\frac{V_c \delta t}{\tau}} \right) + ASI_{t-1} e^{-\frac{V_c \delta t}{\tau}} \quad (3)$$

where t is time; V_c is wind speed.

Air quality index (AQI) is calculated according to technical regulation on ambient air quality index in Table 1 (MEPC, 2012). It is used to assess whether the pollution levels exceed the national criteria at different cities or regions, and provide information on air quality and individual activity advice to public in order to protect public from the health effect of air pollution (Zheng *et al.*, 2014).

$$IAQI_p = \frac{IAQI_{Hi} - IAQI_{Lo}}{BP_{Hi} - BP_{Lo}} (C_p - BP_{Lo}) + IAQI_{Lo} \quad (4)$$

AQI = max

For pollutant p , $IAQI_p$ is individual air quality index; C_p is a mass concentration; BP_{Hi} and BP_{Lo} are the upper and lower limits of the corresponding AQI interval. The AQI is defined

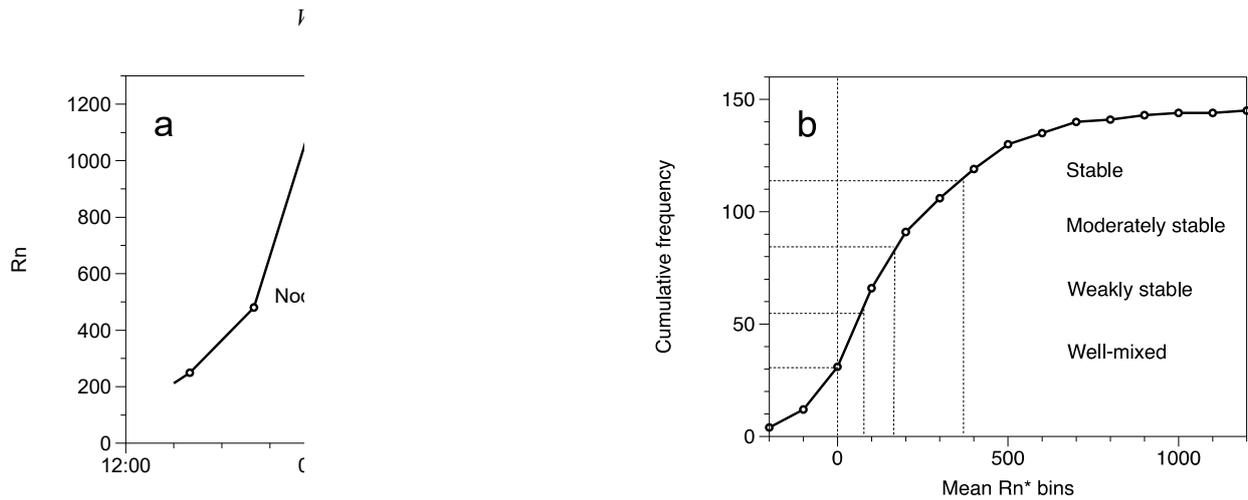


Fig. 2. (a) Diurnal composite I for all of the campaign.

Tab1

Intervals for pollutant concentrations (mg m^{-3})			AQI interval	Air pollution level
SO ₂	NO ₂	PM ₁₀		
[0.000, 0.050]	[0.000, 0.080]	[0.000, 0.050]	0–50	Good
(0.050, 0.150]	(0.080, 0.120]	(0.050, 0.150]	51–100	Moderate
(0.150, 0.800]	(0.120, 0.280]	(0.150, 0.350]	101–200	Unhealthy for sensitive groups
(0.800, 1.600]	(0.280, 0.565]	(0.350, 0.420]	201–300	Unhealthy
(1.600, 2.100]	(0.565, 0.750]	(0.420, 0.500]	301–400	Very unhealthy
(2.100, 2.620]	(0.750, 0.940]	(0.500, 0.600]	401–500	

as the maximum of *IAQI*, and the pollutant responsible for the highest index value is the “Main Pollutant”.

RESULTS AND DISCUSSION

Climatology and ASI of Radon-based Mixing State

Jinhua is located in the Yangtze River Delta (YRD), and the climate in the YRD area is mainly influenced by the East Asian monsoon: warm and wet in summer, cold and dry in winter. Mean monthly wind speeds in Jinhua were low and sustained $1.6\text{--}1.8 \text{ m s}^{-1}$ during the measured period (Fig. 3). The monthly average temperature increased from 13°C in March to 31°C in July, and relative humidity ranged from 67% to 83%, higher in the plum rain season (April, May and June).

Monthly Means and Standard Deviation

The ASI is representative of the combined scavenging capacity for pollutants by wind and precipitation. Near-surface radon measurements, on the other hand, inform only about atmospheric dilution and dispersion, since ambient radon concentration is not significantly impacted by rain. Assuming no large changes in air mass fetch, or the regional radon source function, in principle, the higher the ASI the lower the observed near-surface radon concentration. To investigate how representative modeled ASI values are, both whole-day and nocturnal radon measurements were compared with ASI values. Fig. 4 shows the correlation of ASI with our proxy for radon concentration and the nocturnal mean (2000–0400 h) radon proxy data. Based on whole-day data, ASI has an inverse correlation of power exponents with

radon; in Fig. 4 the $\log(\text{Rn})$ and $\log(\text{Rn}^*)$ are shown and the goodness of fit (R^2) of the linear regression is high, 0.81 for both cases. These results confirm a strong and consistent relationship between measured radon and modeled ASI data.

Using the radon-based stability categories defined in Section 2.2, we sorted diurnal composites of the Jinhua wind speed, temperature, relative humidity records and the calculated ASI of Jinhua (Fig. 5) There are four nocturnal mixing categories: “well-mixed”, “weakly stable”, “moderately stable” and “most stable” and each class is based on 29 separate whole nights. The radon-based classification scheme is able to clearly distinguish changes in mean ASI, wind speed, temperature and relative humidity between the different stability classes.

The most well-mixed nights (Category 1) according to the radon-based scheme, when nocturnal radon accumulation is lowest, consistently exhibit the highest ASI values. They also have the highest relative humidity, lowest temperatures, and high wind speeds that decrease during the morning hours and may be followed by a “near-neutral” day (Chambers *et al.*, 2015) The most stable nights (Category 4) according to the radon-based scheme, when nocturnal radon accumulation is highest, are characterized by the lowest ASI values, wind speeds and relative humidity. The distinction between nocturnal ASI values for Category 1 and 2 days is not always as clear, but this is likely to be largely attributable to a combination of low temporal resolution of radon observations (4-hour instead of hourly) and short nocturnal window (8 hours instead of 10 hours) compared with other applications of the radon-based stability classification

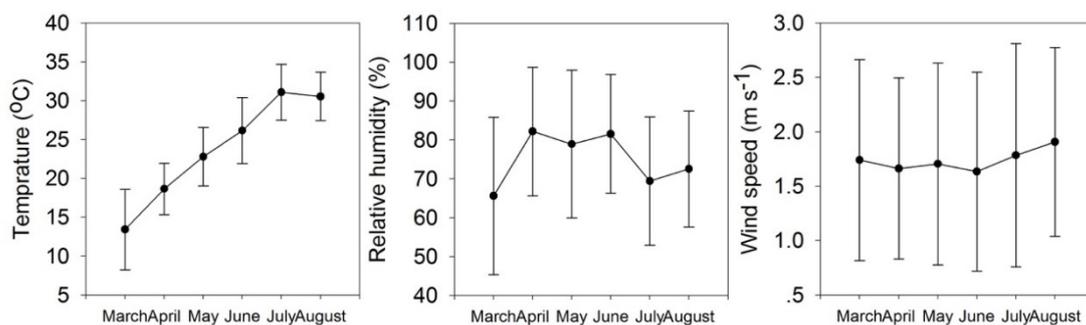


Fig. 3. Summary of Jinhua climatology between March 2016 and August 2016. Filled circles represent the average and bars represent $\pm 1 \sigma$.

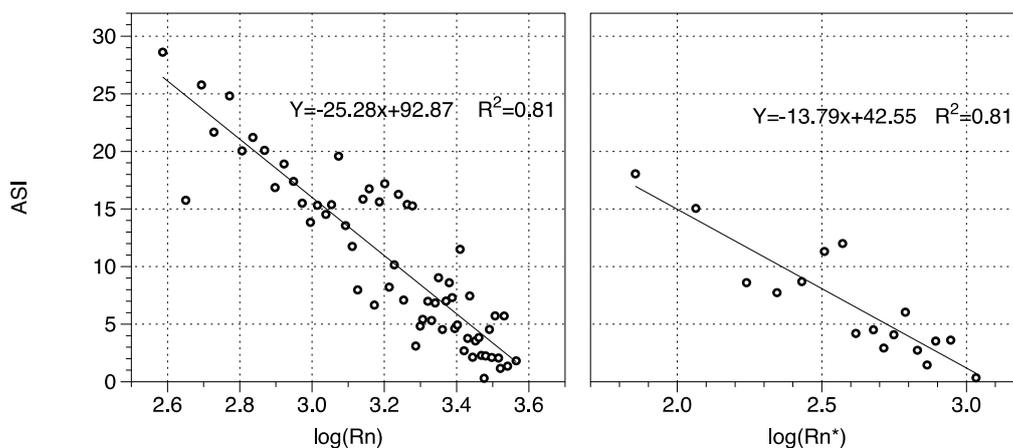


Fig. 4. Correlation of ASI and mean radon concentrations over all day and the period 2000–0400 h during the observation period. 4-hour means of ASI observations of radon have sufficient available data; each point ($\log(Rn)$) represents a mean ASI within a radon data “bin” of 50. (a) Data of a whole day, (b) Subtraction of the average radon concentration during 2000–0400 h ($\log(Rn^*)$) and the value at 2000 h, and the average ASI during 2000–0400 h.

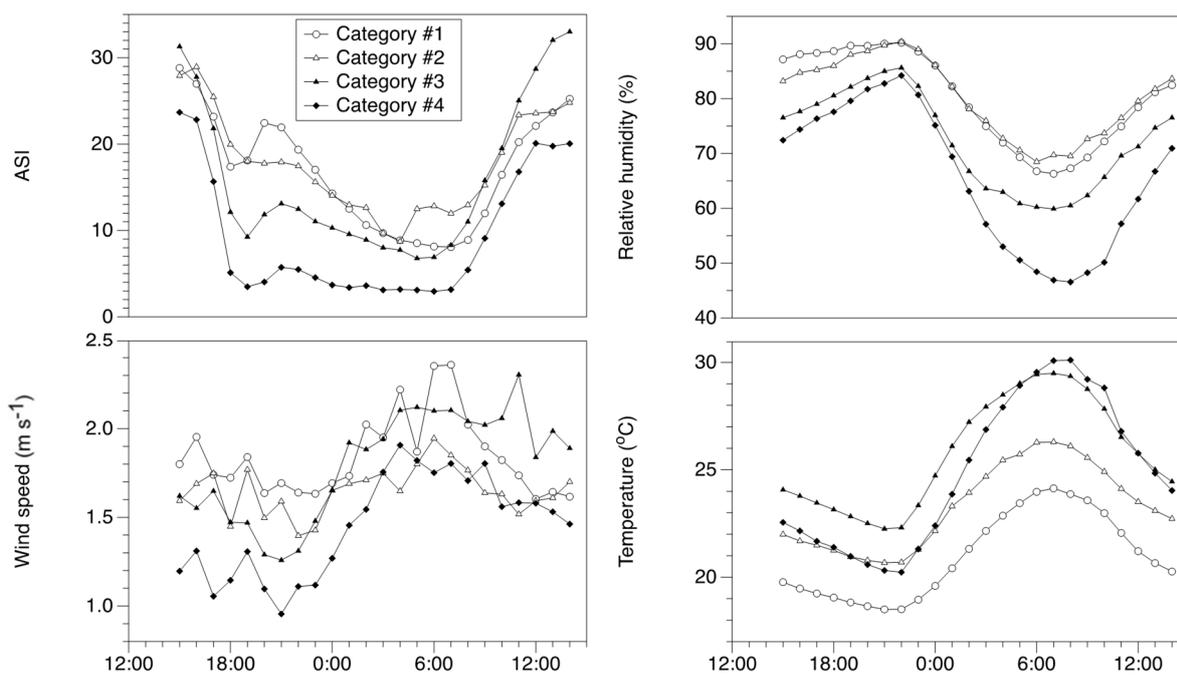


Fig. 5. Hourly mean diurnal composites of ASI, wind speed, temperature and relative humidity as a function of radon-based stability category.

scheme (e.g., Chambers *et al.*, 2015; Williams *et al.*, 2016) There may also be subtle influences of rainfall on ASI values between Category 1 and 2 days not accounted for by the radon-based scheme. The nocturnal (19:00–07:00) mean hourly rainfall are highest at Category 2 (0.43 mm h^{-1}) followed by Category 1 (0.34 mm h^{-1}) and 3 (0.21 mm h^{-1}).

Concentrations of Air Pollutants in Three Stations

We analyzed the hourly (gas phase) and daily average (particulate matter) mass concentrations of the pollutants at three stations. Fig. 6 shows the range and mean concentrations of CO, NO₂, SO₂, O₃-8h and daily PM₁₀, PM_{2.5} during the measurement campaign March 1–August 16, 2016. The O₃, PM₁₀ and PM_{2.5} recorded the most exceedances. O₃-8h values record exceedance of NAAQS Level 1 (Fig. 6). PM₁₀ and PM_{2.5} are the major pollutants, more than 50% exceedance and the median values at three stations were higher than the NAAQS Level 1 50 and 35 $\mu\text{g m}^{-3}$ daily

mean values shown in Fig. 5. On March 6 and March 7, PM₁₀ hourly concentrations were larger than $200 \mu\text{g m}^{-3}$ due to the inference of sandstorm occurred in northwestern China on March 3. And the air quality data during these two days was excluded in the mixing-state influence study in Section 3.4 and 3.5.

The ratios PM_{2.5}/PM₁₀, PM_{2.5}/CO, PM_{2.5}/SO₂, and SO₂/NO₂ were calculated for Station A, Station B and Station C separately, which are dimensionless values. The PM_{2.5}/PM₁₀ ratios were similar at three stations. The ratio of PM_{2.5} to PM₁₀ at three stations are 0.74, 0.67 and 0.68 respectively and similar to the study in Zhejiang Province (0.61–0.70) by Zhao *et al.* (2018) studied during the year 2011 to 2014. PM_{2.5}/CO ratios are consistent 0.05 at three stations, which indicates the secondary formation of PM_{2.5} (CO is regarded as an indicator of primary combustion sources) is comparable in other studies (Song *et al.*, 2017).

PM_{2.5}/SO₂ varies at Station A, B and C (3, 3.24, 4.88) and

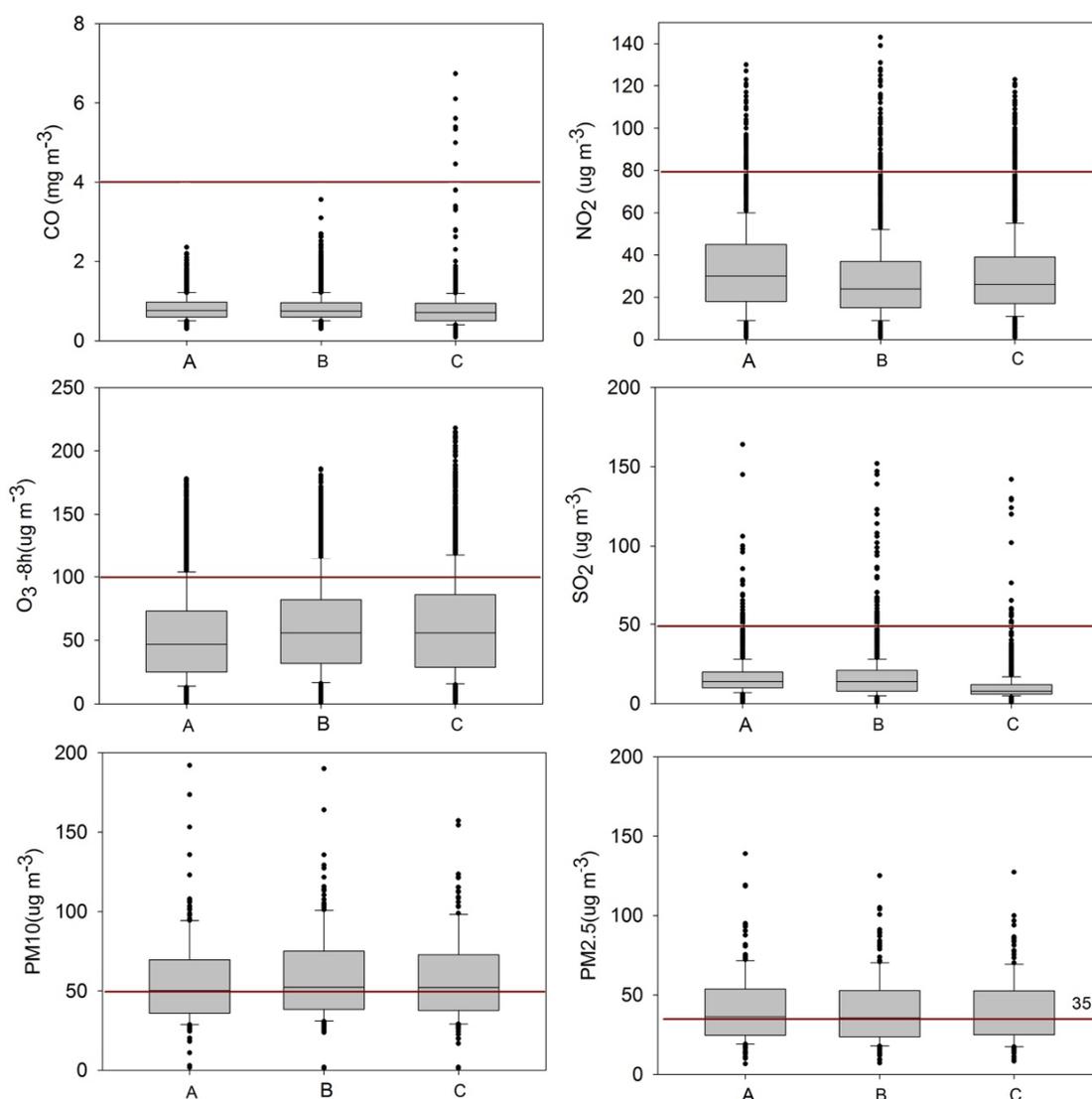


Fig. 6. Box plot of concentrations. For each pollutant, the 25–75% quartiles are drawn using a box. The median is shown with a horizontal line inside the box. The 10th and 90th percentile values are shown with short horizontal lines (“whiskers”). Outliers are shown as circles. The NAAQS Level 1 values are shown in red line for O₃-8h, PM₁₀ and PM_{2.5}.

is higher than the average in Zhejiang ranging from 1.79 to 2.65 for the year from 2011 to 2014 reported by Zhao *et al.* (2018a). SO_2/NO_2 ratios (0.91, 0.80, 0.44) also differ at three stations, indicates that Station A and Station B have more influence by industry emissions (Song *et al.*, 2017). This may be due to the high variability of SO_2 concentration at three stations (Song *et al.*, 2017), since the observation sites may be close to local energy industries.

Effects of Mixing State on Concentrations

In order to investigate the atmospheric stability influences on air pollution level, the diurnal cycles of pollutants O_3 , $\text{PM}_{2.5}$, PM_{10} and AQI were sorted to four category stability classes shown in Fig. 7. Concentrations show similar trends in all three stations, that there are mixed-up concentrations happening between Category 1 and 2, and the large contrast between Category 3 and 4. Heavy pollution episodes appear under the most stable conditions (Category 4) with calm to light gradient wind and less rain, while concentrations of pollutants are lowest under moderately stable conditions (Category 3) with light winds. The diurnal variations are more pronounced with morning and afternoon peaks under Category 3 and 4, which depicts local emissions events. Well-mixed (Category 1) and weakly stable (Category 2)

conditions are associated with higher wind speed, often overcast, and more advection, but the concentrations are often higher than these under Category 4, which indicates that there is a large non-local contribution to pollution in Jinhua, i.e., there is significant pollution advection coming from outside of the city. The combination of local and remote pollution results in higher diurnal values with less dominant morning and evening peaks.

Prediction of AQI by Radon and ASI

AQI prediction is used to indicate the need for pollution prevention activities for the public and to advise the regional emission reduction reaction in serious conditions. It has already been established that nocturnal radon measurements are a reliable means of predicting haze (visibility) problems the following morning (Wang *et al.*, 2016). We used radon and ASI to predict rush hour AQI (0800–1000 h) by simple predictive models (Fig. 8), which can be set up for automatic forecast system. The mean radon observations and ASI between 2000–0400 h in the night before were used to predict the next-morning AQI. ASI shows reliability ($R^2 = 0.67$) for AQI prediction referring to AQI bins of 10. The air pollution level can be predicted as “good” when $\text{ASI} > 10$, “moderate” when $5 < \text{ASI} \leq 10$, “unhealthy” when $\text{ASI} \leq 5$

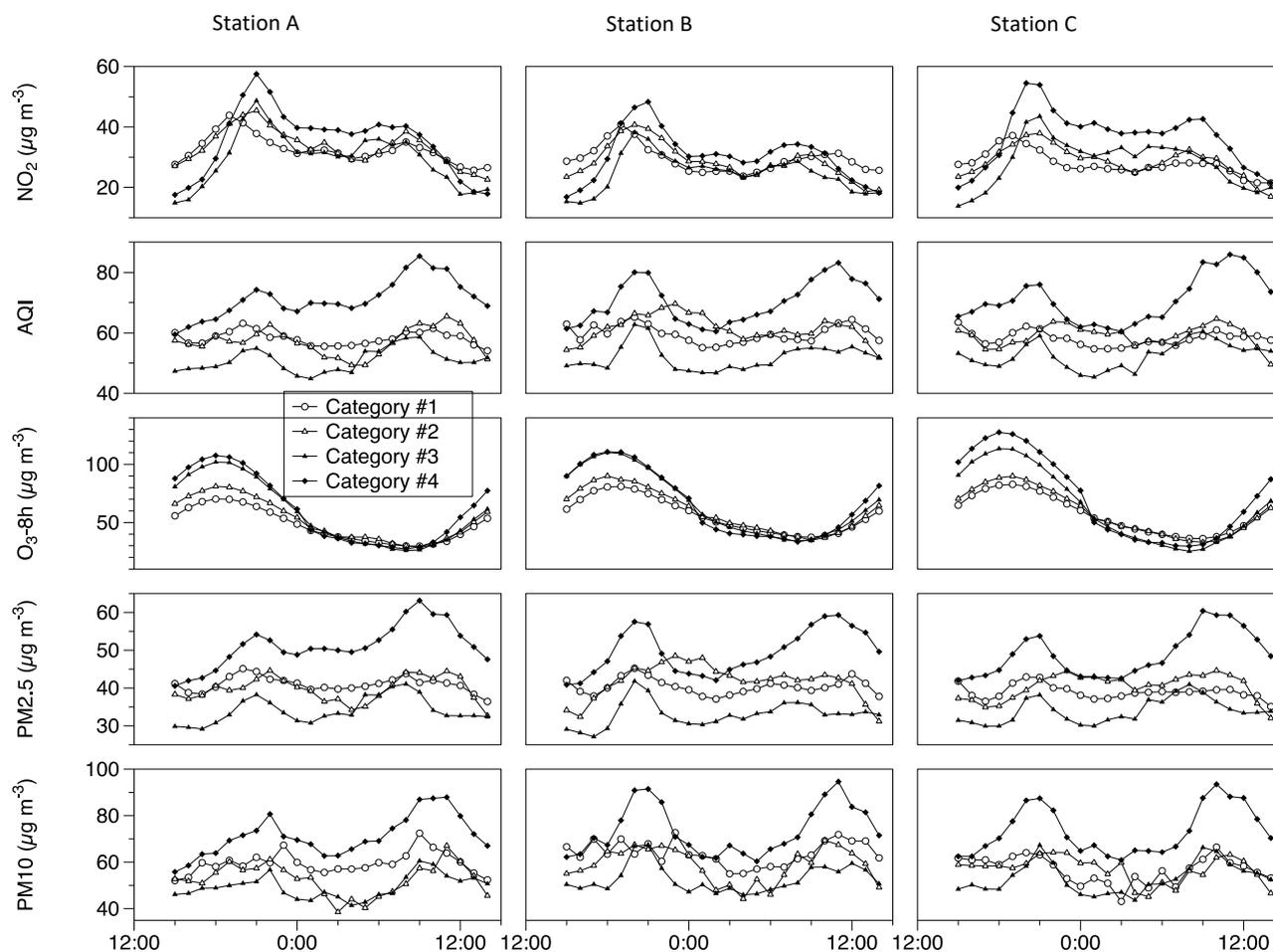


Fig. 7. The influence of nocturnal atmospheric stability on the diurnal cycles of select pollutants and AQI at Stations A, B and C in the whole measurement period.

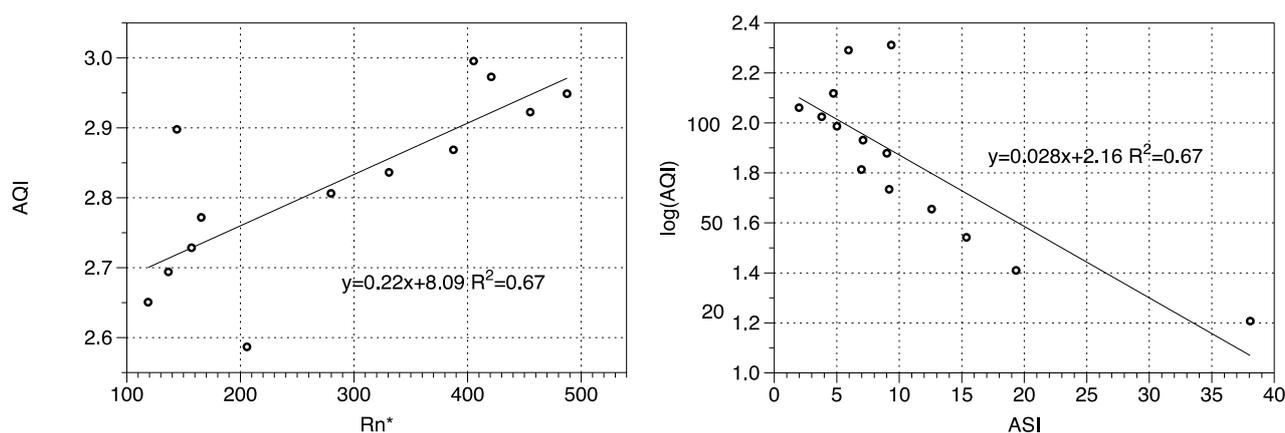


Fig. 8. Predictive models of morning AQI (0800–1000 h) based on nocturnal radon (left) and ASI (right), excluding heavy rainfall events. 4-hour means of ASI observations and radon have sufficient available data, each point representing a mean ASI within an AQI data “bin” of 10. The log(AQI), and AQI are shown at y-axis legend.

in the city of Jinhua. When there is ASI moderate class, sometimes AQI can be higher than 100, which have to be analyzed combining individual meteorological condition analysis. City clusters in the YRD recorded high similarity of synoptic phenomena, thus these values may be similar for cities having close climatic conditions and difference between cities belongs to diverse climatic conditions.

The limitation of using radon measurement to predict AQI is the prediction time is relatively short, and time latency is too short for taking any reaction in case heavy pollution events will occur. ASI can be predicted by the WRF model. Then AQI can be forecasted using the ASI from the model for several days in advance, or even several weeks depending on the ASI prediction period and precision.

CONCLUSIONS

A modified radon-based scheme for characterizing nocturnal atmospheric stability was used to assign stability categories to observation data collected over six months at three sites in Jinhua, China, in conjunction with the results from previous studies conducted in Lanzhou, China (Chambers *et al.*, 2015; Wang *et al.*, 2016). The near-surface atmospheric radon was measured from March to August 2016 with an SM200 stability monitor, and the pollutant concentrations (PM_{2.5}, PM₁₀, SO₂, NO₂, CO, and O₃) and air quality index (AQI) values recorded at three stations were analyzed according to the four-category nocturnal mixing scheme. The concentrations and ratios of the pollutants exhibited differences between the various stability classes, which were primarily attributable to a combination of stability-related changes in the mixing depth, the air mass fetch, and the proximity to major pollution sources. The atmospheric self-cleaning ability index (ASI) was calculated, and the atmospheric radon measurements were found to be consistently representative of the atmospheric mixing state. Thus, we propose a simple model based on the ASI for predicting the AQI during the morning rush hour.

Our study indicates that near-surface radon measurements provide a reliable tool for evaluating air quality and reducing

pollution by considering the meteorological conditions that are most conducive to severe pollution episodes. In combination with a reliable weather/climate forecasting model, the proposed air quality prediction model can be extended to generate forecasts weeks and months in advance, thereby reducing the public exposure to heavy air pollution.

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