Noble Gas Composition in Rainwater and Associated Weather Patterns

Supplementary Text 1: Detailed weather data description for individual samples

1. Introduction to weather products

For each of the precipitation events associated with our sampling collection, multiple weather products were analyzed to provide a detailed characterization of each one of these events. Detailed weather information for each sample include: a) synoptic scale weather patterns such as movement of cold/warm fronts and location of high/low pressure ridges/troughs; b) precipitation characteristics such as rainfall intensity (light, moderate, heavy rainfall), presence or absence of thunder, hail, fog; and c) probable condensation altitudes.

Synoptic scale weather features were obtained using surface analyses maps based on station observations issued every three hours by the Hydrometeorological Prediction Center (archived at http://www.hpc.ncep.noaa.gov/html/sfc_archive.shtml). Figures S3, S4 and S5 show examples of such maps corresponding to rainfall events during collection of samples nr1, 6, and the two samples 10-1 and 10-2. Similar maps are used to describe weather features in detail for each rainfall event during collection of all samples (cf., section 2 below). Description of various fronts and boundaries shown on the maps are available at the National Weather Service glossary (http://www.nws.noaa.gov/glossary/).

Synoptic scale weather pattern descriptions obtained through surface analysis maps were crosschecked with area forecast discussions issued by the National Weather Service for the Detroit/Pontiac region (archived at http://mesonet.agron.iastate.edu/wx/afos/list.phtml). Text S2 provides examples of unedited area forecast discussions that are most up to date prior to precipitation events of samples nr1, 2, 6 and 12. Precipitation characteristics for each sampling event were derived from available surface weather station observations. All weather station records were obtained from the Integrated Surface Hourly database published by the National Climatic Data Center (NCDC; http://gis.ncdc.noaa.gov/web/ish.html). Because an hourly weather station is not available in Milan, MI, the closest available weather stations (cf. Fig. S6) located in Ann Arbor (~15 kms; NCDC WBAN station number: 725374), Willow Run (~19 kms; NCDC WBAN station number: 725376) and Custer (~27 kms; NCDC WBAN station number: 725418) were used to derive precipitation characteristics in Milan. Individual weather station records used for each sampling event are shown in Table S1. All weather records relevant to a particular precipitation event are included in Table S2. Data description for each column in Table S2 including units, abbreviations and weather codes are provided in detail by NCDC (http://hurricane.ncdc.noaa.gov/cdo/3505doc.txt). It should be noted that weather data from a particular weather station was utilized only after ensuring that the specific storm passed through both the sampling location and appropriate weather station at the precise time of sampling by analyzing archived Doppler radar images (available every 5 minutes) for southeast Michigan (NEXRAD available at...
http://www.ncdc.noaa.gov/oa/radar/radarresources.html). Figure S6 shows an example of a Doppler radar image for the precipitation event during collection of sample nr2. Similar images were obtained for all other samples using the same archive. Condensation altitudes for each sample were estimated from available weather balloon (sounding) data at Detroit, MI (42.695°N, -83.467°W; Fig. S1). Similar to weather station records, sounding data from Detroit for a particular rain event was utilized only after ensuring that the specific storm passed through both the sampling location and Detroit (~80 kms and ~70 kms from Milan and Ann Arbor, MI, respectively) at the precise time of sampling by analyzing archived Doppler radar images for southeast Michigan. Sounding data is available twice daily (at 0 and 12 UTC (Coordinated Universal Time)) from the NOAA/ESRL Radiosonde database (esrl.noaa.gov/raobs) and is plotted in the form of SkewT-logP plots (http://RUCsounding.noaa.gov), which is a vertical snapshot of temperature, dew point and winds above a point on Earth. Further information about radiosonde (weather balloon) data and SkewT-log P plots is available at the Federal Meteorological handbook at www.ofcm.gov/fmh3/text/default.htm. Figure S7 shows examples of SkewT-log P plots corresponding to samples nr1, 6, 11 and 12. Probable condensation altitudes can be estimated as those altitudes in the SkewT-log P plot at which measured temperature (red lines, Fig. S7) is similar to the dew point (blue line, Fig. S7). This is discussed in detail in section 2.

Weather data descriptions obtained by simultaneously analyzing all the above weather products for individual rain events during collection of each sample are provided below. As mentioned above, weather products are available only at specific hours that may or may not necessarily correspond to the precise time of our sampling event (Table S1). However, weather descriptions provided below utilize the most recently available weather products prior to the sampling event and represents our best attempt at describing the weather patterns at the time of sampling. Nevertheless, synoptic weather patterns and precipitation characteristics collectively derived clearly seem to differ vastly between mass independent (nr1, 8, 12, 14) and mass dependent samples as described below. A brief summary comparing the distinct weather patterns observed for mass dependent and mass-independent samples is also provided (Section 4).

2. Detailed description of weather information for Mass-Independent samples

2.1. Sample nr1

The surface analysis map (Fig. S3) indicates the presence of a low-pressure trough in the Ohio valley producing precipitation in southeast Michigan. Similar synoptic scale weather patterns were identified in the area forecast discussion (cf. Text S2). Surface weather records at Ann Arbor indicate the presence of mist/fog, light to moderate rain and low cloud ceiling heights (~300m) through both manual and automated observations between 15:12 UTC and 15:53 UTC when the sample was collected (Table S2, Figure S8). Available temperature and dew point sounding at 12 UTC visualized in the SkewT-log P plot (Fig. S7) indicates that condensation can occur between ~300m and ~3kms (1000ft and 10,000ft) and probably at higher altitudes (e.g., 3.6kms or ~12,000ft) where air temperatures are below 0°C. This suggests that in addition to the low clouds/fog observed through surface observations, condensation originating as ice/ice melt from higher condensation altitudes are definitely likely.
2.2. Sample nr8

The surface analysis map at 18 UTC indicates the presence of a low-pressure trough across the lower peninsula of Michigan. Between 18:34 UTC and 19:34 UTC when nr8 was collected, surface weather observations at Custer weather station indicate the presence of intermittent rain and mist (cf. Table S2). The presence of fog/mist and low altitude clouds was also predicted in the area forecast discussion. SkewT-log P plots of temperature and dew point sounding data at 12 UTC and 24 UTC on August 6th, 2011 indicates the probability of multiple condensation levels in addition to the presence of low altitude clouds. Some of these condensation levels occur at altitudes where temperatures are below 0°C suggesting that condensation originating as ice/ice melt are likely.

2.3. Sample nr12

Area forecast discussion and surface analyses maps at 6 UTC and 9 UTC show that precipitation collected at 7:50 UTC is associated with an upper level low pressure system ahead of a warm front moving in to southern Michigan from the southwest. Surface weather station at Custer observed light continuous rain at 7:53 UTC. While the presence of fog was not reported at this weather station, area forecast discussions had predicted the presence of fog (cf. Text S2). In addition, surface temperature and dew points were the same during the period of sample collection indicating the presence of low-altitude clouds/fog (cf. Fig. S8). SkewT-log P plots of temperature and dew point sounding data at 0 UTC and 12 UTC also indicate that low altitude clouds are likely (cf. Fig. S7). In addition, SkewT-log P plots also indicate condensation levels at higher altitudes (~4.5 kms or ~15,000 ft) where temperatures are below 0°C suggesting that condensation originating as ice/ice melt is likely.

2.4. Sample nr14

Similar to other samples with mass-independent patterns, both the surface analysis map at 15 UTC and the area forecast discussion suggest that this precipitation event was due to the presence of a low pressure trough south of Michigan in the Ohio Valley. Surface weather records at the Ann Arbor weather station at 15:10 UTC indicates continuous, moderate rain with the presence of mist identified both manually and through automated observations. Low cloud ceiling heights (less than 300m) were observed at the Ann Arbor weather station and previously predicted in the area forecast discussions. However, sounding data is not available for this precipitation event because NEXRAD images do not indicate that the storm passed through both Detroit and Milan at the time of sample collection. Thus, likely condensation altitudes could not be predicted for this sample.

3. Detailed description of weather for mass dependent samples

3.1. Sample nr2

Area forecast discussion and NEXRAD images (Fig. S6) for this precipitation event indicate the occurrence of an isolated thunderstorm due to lake breeze interactions. This isolated thunderstorm misses all available weather stations at the time of sample collection.
collection (Fig. S6). Thus, precipitation characteristics and condensation altitudes could not be derived for this sample.

3.2. Samples nr3-1, nr3-2 and nr4-1, nr4-2

Area forecast discussions predict the occurrence of thunderstorm clusters during the eastward movement of a mesoscale convective complex across southern Michigan during sample collection of nr3-1, 3-2 followed by a second upper level disturbance igniting additional thundershowers during collection of nr4-1, 4-2. While the storm passes through Custer weather station, cloud ceiling height and precipitation characteristics observations were not recorded during the time of sample collection of nr3-1, -2 (cf. Table S1, S2). However, during sample collection of nr4-1, 4-2, the Ann Arbor weather station (cf. Table S2), through which the storm passes, recorded moderate to heavy rain with showers of hail between 21:53 UTC and 22:05 UTC. In addition, SkewT-log P plot at 0 UTC corresponding to nr4-1, 4-2 suggest the presence of multiple condensation levels including at altitude of ~4.5km (~15,000 ft) where temperatures are below 0˚C suggesting that condensation originating as ice/ice melt is likely.

3.3. Samples nr5-1 and nr5-2

Surface analyses and area forecast discussions indicate initially a stationary front followed by an advancing warm front that moves east from northern Illinois to southern Michigan bringing with it a thunderstorm complex. Precipitation characteristics recorded at the Ann Arbor weather station initially indicate continuous, moderate rain and mist followed by heavy rain, thundershowers and hail showers (Table S2). The heavy rain is also captured by the Doppler radar images for these precipitation events. SkewT-log P plot created with temperature and dew point measurements from weather balloons at 0 UTC on July 28th, 2011 indicate that condensation altitudes up to ~4.5km (~15000 ft) are likely. Because temperatures are ~0˚C at these altitudes, condensation originating as ice/ice melt is probable.

3.4. Sample nr6

Both the surface analysis map (Fig. S4) and area forecast discussion suggest that this precipitation event occurs due to convection along a stationary front that is positioned in a southwest-northeast direction across the lower peninsula of Michigan. Area forecast discussions further suggest the likelihood of numerous thunderstorms in southeast Michigan. Indeed, surface weather observation at Ann Arbor indicates moderate to heavy rain with showers of hail between 8:13 UTC and 8:51 UTC when sample nr6 was collected (Table S2). In addition, SkewT-log P plot at 0 UTC and 12 UTC (Fig. S7) suggest the presence of multiple condensation levels including altitudes of 1.2km, 2.1km, 4.5km and 6km (or 4000ft, 7000ft, ~15,000ft and 20,000ft). Measured temperatures are below 0˚C above ~4.5 km (red line, Fig. S7) suggesting that condensation originating as ice/ice melt is likely.

3.5. Sample nr7

Surface analysis and area forecast discussions suggest heavy rainfall and thunderstorms produced by instability in southern Michigan as indicated by a stationary front. The presence of heavy rain at the time of sample collection is also indicated by
Doppler radar images and confirmed by surface weather observations at the Ann Arbor weather station which record heavy rain with hail showers at 7:13 UTC, close to the sample collection instant (Table S2). In addition, cloud base heights recorded at these very same surface weather stations are high (between 1.2km and 2.4km or 4200ft and 8000ft) and suggest multiple condensation levels. Multiple condensation levels at ~1.5km, 3km, 4.5km and 5.4km (or 5000ft, 10,000ft, 15,000ft and 18,000ft) are also evident through SkewT-log P plots at 0 UTC. Condensation at altitudes higher than ~4.5kms where temperatures are below 0˚C suggest that precipitation originating as ice/ice melt is likely.

3.6. Samples nr9 and nr10-1, 10-2

Surface analysis indicates the movement of a warm front across southern Michigan in a northeast direction producing strong upper level moisture advection generating heavy rain and thunderstorms as also described in the area forecast discussions for nr9. This is followed by a cold front associated with a low pressure system over Ontario, which sweeps across southern Michigan from west to east bringing with it showers and thunderstorms during which nr10-1 and 10-2 are collected (Fig. S5). Surface weather observations in Ann Arbor also indicate moderate to heavy rainfall with thunderstorms and hail showers between 9:33 UTC and 9:36 UTC when nr9 was collected (Table S2). While thunder with rain is reported in the surface analysis map ahead of the cold front during nr10-1 and nr10-2 sample collection, surface weather data is not available at Willow Run station, through which this particular storm passes, to confirm this. However, we have physically observed thunderstorms and hail during sample collection of nr10-1 and 10-2. In addition, we observed a significant drop in temperature (2.8˚C) at the collection site between sampling of 10-1 and 10-2 suggesting that the cold front passed through Milan during that time interval. Sounding plots for both samples indicate multiple condensation level where temperatures are below 0˚C and suggests the likelihood of precipitation originating as ice/ice melt.

3.7. Sample nr11

Surface analysis map indicates a stationary front slowly moving across southern Michigan from north to south associated with a mid level storm rotating across northern Ohio bringing with it rain and thunderstorms as indicated in the area forecast discussion. Surface weather station at Ann Arbor does not report any particular precipitation characteristic during sample collection (Table S2). Similar to all other samples, multiple condensation levels below 0˚C are possible as indicated by SkewT-log P plots (Fig. S7) suggesting the likelihood of precipitation starting as ice.

3.8. Sample nr13

Surface analyses maps indicate a stationary front positioned across southern Michigan, which then moves as a cold front across the Lower Peninsula. Area forecast discussion indicates that this will bring clusters of thunderstorms to southeast Michigan. These are confirmed by surface weather observations at Ann Arbor, which record heavy rain, thunderstorms and showers of hail (Table S2). In addition, sounding plots point to multiple condensation levels and the presence of condensation altitudes below 0˚C.
4. **Summary of weather analyses for mass-independent and mass-dependent samples**

Our detailed weather analyses above indicate that all rain samples with mass-independent patterns were collected during precipitation due to the presence of a low-pressure system in the area. Weather records for all samples with mass-independent patterns, without exception, indicate that corresponding precipitation events can be characterized by the presence of fog/mist, light to moderate rain and low cloud ceiling heights (~300m). By contrast, samples with mass-dependent patterns were collected during energetic thunderstorms due either to a stationary front (e.g., nr6), cold front (e.g., nr10-1/10-2) or warm front (e.g., nr9). Surface weather stations confirm this by recording heavy rain, thunderstorms and showers of hail during the time of sample collection. Of particular significance, is the fact that except for some initial fog activity during the approach of the thunderstorm during collection of sample nr5-1, no other surface weather station records the presence of fog or low-level clouds during collection of mass-dependent samples. In addition, all samples, including those displaying mass-dependent and mass-independent patterns indicate the possibility of multiple condensation levels, including altitudes where air temperature is below 0°C. These results suggest the likelihood of precipitation starting as ice in southeast Michigan, a finding that was also previously suggested by *Bernstein et al.* [2007].

**References**