Reply to reviewer #2: Interactive comment on "Continuous online-monitoring of Ice Nucleating Particles: development of the automated Horizontal Ice Nucleation Chamber (HINC-Auto)" by Cyril Brunner and Zamin A. Kanji.

Reviewer comments are reproduced in **bold** and author responses in normal typeface; extracts from the original manuscript are presented in *red italic*, and from the revised manuscript in *blue italic*.

In this paper authors described the HINC-Auto, which is automated version of HINC. They describe the technical setup and validation experiments. CFD modeling is also performed. The chamber was deployed in the field and ran for 90 days. The paper is well written, and I recommend publication after following minor points are addressed.

We would like to thank the reviewer for the compliments and valuable comments and address the concerns individually below.

Line 104 -105: Is flow rate affects the buoyancy? Can you have larger gradient but smaller flow rate?

From our understanding, a larger temperature gradient between the warm and cold wall would exacerbate the buoyancy at the warm wall. The difference in buoyancy will introduce shear within the fluid. A smaller flowrate will lead to less shear within the fluid. Thus, increasing the temperature gradient while decreasing the flow rate in order to maintain the same shear is probable. Exactly how the flow rate would influence the buoyancy would require complex 3D fluid dynamics simulations. However, we note that this aspect pertains to vertical chambers, which is not the topic of this paper.

Figure 2: For completeness label the vacuum pump. Currently, the output air is recirculated – closed loop configuration. What is the need of MFC after OPC?

We agree with the reviewer's comment and incorporated the proposition in Figure 2b:

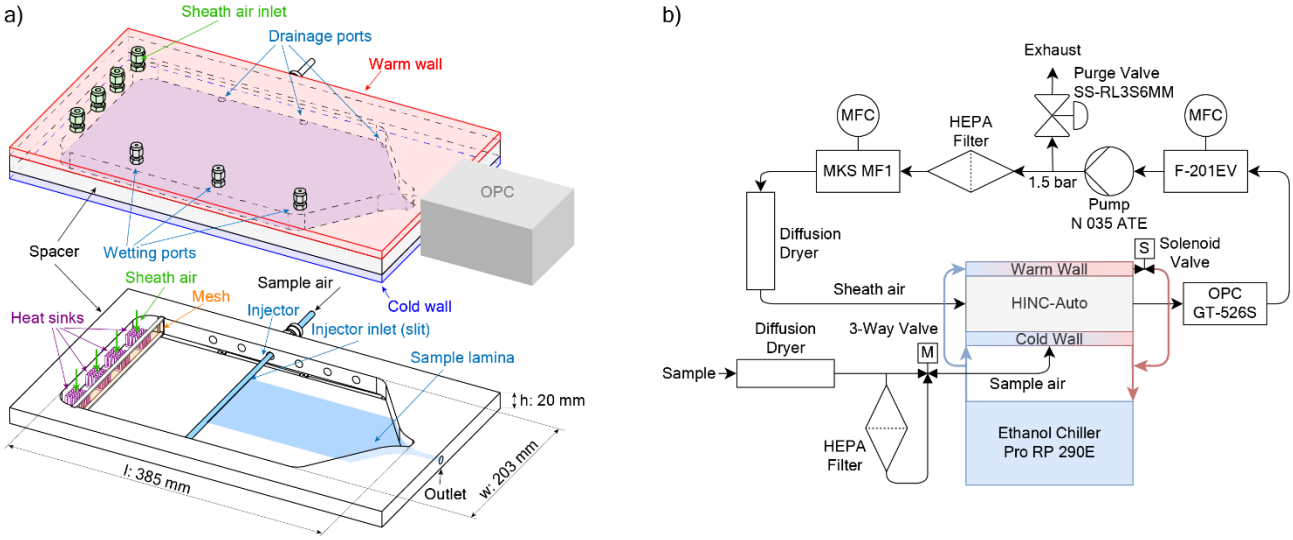

Figure 2.

The MFC after the OPC is needed to maintain a chamber outlet flow rate of 2.83 std L min^{-1}. Controlling both outlet and sheath flow rate allows to indirectly control the sample flow rate without the need of a MCF in the sample flow: chamber outlet flow rate = sheath flow rate + sample flow rate. This is stated in line 147: The sample air flow rate of 0.283 std L min^{-1} results from the difference of the volume flow exiting the chamber through the OPC and the sheath air directed into the chamber.

Line 123: How many layers of filter paper were used.

One layer of filter paper was used. We changed line 124 (revised manuscript) as follows:
The inner metal walls are each covered with one layer of self-adhering borosilicate glass microfibre filter paper (PALL 66217, 1 µm, 8x10") which is wetted with water and acts as reservoir for water vapor in order to create ice and/or water (super)saturation.

Section 2.1: A plot showing the time series of temperature, RH, and droplet diameter would help to understand the operation of the chamber. This plot can include droplet growth, rewetting, and INP measurement periods.

We thank the reviewer for the comment. We already include a time series of RH in Figure 4 and A4 and discuss droplet/ ice crystal growth in section 2.4. Showing one time series with temperature, supersaturation and hydrometeor growth is somewhat arbitrary, as the plot substantially changes for each set temperature, supersaturation, ambient pressure and injector position. Section 2.1 provides just background information over CFDCs without having introduced any concepts like injector position or rewetting/ INP measurement periods. Therefore, we think it is too premature to show Figure 4, A4 or an adaption including hydrometeor growth at this point i.e. in section 2.1.

Line 189: CDF or CFD?

We meant, CFD – thanks for catching that. We changed line 191 (revised manuscript) accordingly:

The 3D CFD simulation revealed for the supersaturation to need a substantial part of the chamber length to equilibrium to the set conditions.

Section 2.4: Regarding particle losses. Is there any size-dependent transmission curve established? Any particular reason why >3 um cannot be transmitted? Because of the presence of any upstream impactor?

We measured the particle counts using an OPC during a Saharan dust event at the JFJ on February 9th, 2020. During the dust event, also particles with an optical diameter of 4.0 µm were present. A 15-minutes cumulative measurement was performed outside next to the total aerosol inlet, at the sample line, where normally the injector of HINC-Auto would be connected (i.e. after the dryer and valve), and at the outlet of the chamber, while both chamber walls were held at 20 °C. Table 1RC2 shows the transmission fraction of the ambient particles. No upstream impactor was used. We believe the horizontally oriented injector tubing upstream of HINC-Auto and injector within HINC-Auto allow for sedimentation, which limits their transmission rate.

	1.0 µm	1.5 µm	2.0 µm	3.0 µm	4.0 µm
Outside, next to Total Inlet	100%	100%	100%	100%	100%
After Dryer + Valve	72%	59%	40%	21%	14%
After Chamber Walls at 20°C	69%	58%	33%	0%	0%

Table 1RC2. Transmission fraction of ambient particles during a Saharan Dust event at the JFJ on February 9th, 2020.

Furthermore, we do not believe this transmission efficiency affects our results in a significant manner since we reach an AF = 0.98 for sampling ambient particles at JFJ (see figure 6, revised manuscript).

How droplet diffusion growth calculations are performed? Can this equation be added to the appendix? It is not clear why d0 = 2 um used. A typical size-distribution at JFJ can be shown to understand what is d0.

We thank the reviewer for highlighting the missing reference. The diffusional growth calculations are according to Rogers and Yau (1989). We added the citation to line 226 (revised manuscript):

Diffusional growth calculations (Rogers and Yau, 1989) … (See continuation of text after the next **reviewer comment**)

Furthermore, we added the following passage to the appendix; line 545 (revised manuscript):

Diffusional growth is calculated according to Rogers and Yau (1989) with the latent heat of sublimation of ice, and latent heat of vaporization of supercooled water according to Murphy and Koop (2005). T and S are variable and feed in from the 2D diffusion model corresponding to the particle's current horizontal and vertical position within the chamber. The diffusional growth of the hydrometeors assumes activation when saturation with respect to ice or water is exceeded.

d_0 = 2µm was used, as it is the largest observed particle diameter able to transmit through the tubing and HINC-Auto at the JFJ. We changed line 221 (revised manuscript) as follows:

The transmission fraction of ambient particles ≥ 2 µm through the tubing and the dry chamber (both walls held at 293 K) on the JFJ is 33%. No ambient particles ≥ 3 µm were transmitted. Therefore, to assess the maximum size of droplets in the following diffusional growth calculations a maximum initial radius of d_0 = 2 µm is used.

How ice crystals are distinguished from water droplets? The description on line 238- 240 is not clear. It is also mentioned that d = 0.2 um grew to 4.57 um. This indicates droplets and ice crystals co -exist. Please clarify.

This is a valid comment, for which we like to thank the reviewer. Stating first the maximum diameters assuming a constant *T* and S_w, then saying, that this is not how particles experience *T* and S_w within the chamber, as *T* and S_w need to equilibrate, and then stating the maximum diameters with variable, equilibrating *T* and S_w is confusing for the reader. Therefore, we altered lines 224-249 (revised manuscript) to explain this more clearly for constant *T* and S_w, and then quantitatively stating the maximum diameters with variable, equilibrating *T* and S_w:

Diffusional growth calculations (Rogers and Yau, 1989) with set fixed T (e.g. constant at 243 K) and S_w (e.g. constant at 1.04) conditions overestimate the final hydrometeor size at the chamber exit since the calculation assumes a constant supersaturation to be maintained for the entire time the particle passes through the chamber. In reality, the saturation in the particle stream needs to equilibrate to the set conditions, thus, the particles are exposed to a lower saturation for the first few seconds (see Figure A4). The 2D diffusion model provides an estimate of the real T and S_w when using the diffusional growth calculations by Rogers and Yau (1989). For an initial diameter of d_0 = 2 µm, liquid droplets are calculated to grow to a maximum size of d_{liq} = 3.31 µm (Zurich, 965 hPa, τ = 9.1 s) and d_{liq} =2.36 µm (JFJ, 645 hPa, τ = 6.1 s). Measurements of a highly hygroscopic aerosol, ammonium nitrate with an initial mobility diameter of d_m = 200 nm (for the sample preparation see Section 3.2) show the onset of cloud droplets (no ice crystals since T > 235 K) in the ≥ 3 µm-size bin at S_w = 1.038, as seen in Figure 6a, and support the calculated maximum size of 3.31 µm at Sw = 1.04. The impact on the final diameter for an initial size of $d_{0\ 0.2}$ = 200 nm compared to $d_{0\ 2.0}$ = 2 µm is 0.63 µm ($d_{liq\ 2.0}$ = 3.31 µm vs. $d_{liq\ 0.2}$ = 2.68 µm at 965 hPa and τ = 9.1 s). If the INPs activate as soon as ice saturation is exceeded, the ice crystals grow to $d_{ice\ 2.0}$ = 7.77 µm, $d_{ice\ 0.2}$ = 7.51 µm at 965 hPa and τ = 9.1 s and $d_{ice\ 2.0}$ = 7.66 µm (JFJ, 645 hPa, τ = 6.1 s). Therefore, for experiments performed at T = 243 K and Sw = 1.04, all particles detected in the size bin ≥ 4 µm are considered to be ice crystals formed on INPs. Figure 6b shows a measured activated fraction (AF) curve of ambient air on the JFJ during a high INP concentration period (7:05 22. March 2020, UTC). AF is the ratio of all particles, that are detected in the indicated size bin, to all sampled particles, measured with a CPC within the sample flow. The onset of cloud droplets in the ≥ 0.3 µm size bin exactly at Sw = 1 demonstrates the accuracy of HINC-Auto. At S_w = 1.13 an observed steep increase in AF in the ≥ 3 µm-OPC size bin indicates droplets only grew larger than 3 µm at

this Sw. Compared to the ammonium nitrate measurements performed at Zurich, a delayed activation is observed. This is expected because of the decrease in ambient pressure, which results in shorter residence times, and the much lower hygroscopicity of ambient particles at the JFJ compared to ammonium nitrate. The signal visible in the ≥ 4 µm-OPC size bin comes from INPs, which nucleate and grow to ice crystals at S_w ≥ 1.028 (Si ≥ 1.378). This validates the calculations above that at S_w = 1.04 droplets cannot grow to sizes ≥ 4 µm but ice crystals can, thus supporting the use of the ≥ 4 µm size bin to detect ice crystals.

Equation 1: Define the term [LOD] in the RHS or it is saying the units of LOD are in std Lmin-1 – if so move the units to another line. It is not clear how this equation is formulated. The number '60' in the numerator is confusing. Is this number not used to convert the 'V' into std L per sec from std L per min? If so then units of 'V' should be revised. What are the units of 'BG_counts' parameter?

The RHS term refers to the units of the LOD. We deleted the expression *[LOD] = std L min^{-1}*. Supported by the reviewer comment about **Section 3.3** (see the comment later on) we now show how the INP concentration is calculated. Therefore, some variables (ΣBG counts, $\Sigma N_{BG\ samples}$, V and t_{OPC}) are introduced there and not following the calculation of the LOD in equation (2). We changed line 256 (revised manuscript) as following:

The INP concentration is calculated as follows:

$$INP\ concentration = \left(\frac{\sum INP\ counts}{\sum N_{INP\ samples}} - \frac{\sum BG\ counts}{\sum N_{BG\ samples}}\right)\frac{1}{Vt_{OPC}} \quad (1)$$

where ΣINP counts is the sum of all counts (particle number) in the ≥ 4 µm OPC size bin during the INP measurement, $\Sigma N_{INP\ samples}$ is the total number of OPC intervals during the INP measurement, ΣBG counts is the sum of the background counts (particle number) in the ≥ 4 µm OPC size bin before and after the INP measurement while sampling through a particle filter, $\Sigma N_{BG\ samples}$ is the total number of background OPC intervals before and after the INP measurement, t_{OPC} is the duration of each OPC interval in minutes (here 5 sec, thus 5/60 min), and V is the sample flow rate, here V = 0.283 std L min^{-1}. As the volume flow through the OPC is controlled by the MFC in std L min^{-1}, the resulting INP concentration is INP std L^{-1}.

The limit of detection (LOD) is calculated as follows:

$$LOD = \frac{\sqrt{\sum BG\ counts}}{\sum N_{BG\ samples}}\frac{~~60~~1}{Vt_{OPC}} \qquad ~~[LOD] = \text{std L min}^{-1}~~ \quad ~~(1)~~\ (2)$$

where the LOD is in std L^{-1}. If over a period of 120 OPC background sampling intervals with a duration of 5 seconds each a total of 3 counts where detected in the ≥ 4 µm OPC size bin, the LOD would be = 0.612 std L min^{-1} ($\Sigma N_{BG\ samples}$ = 120, t_{OPC} = 0.083 min, ΣBG counts = 3, V = 0.283 std L min^{-1}). The stated LOD provides a 62.3% (1 σ) confidence interval.

Concerning the number '60': it was used to convert the OPC duration from seconds to minutes. We changed Equation 1 by removing the '60' and changing the text in line 262 (revised manuscript) from seconds to minutes.

'BG_counts' are just particles where each count represents a particle, so the unit could be particles or number (#). To clarify, we added *"(particle number)"* where we defined background counts in the OPC after equation 1. See line 258 (revised manuscript).

Furthermore, we realized we stated that V is the flow rate through the OPC, which is false. It should be the sample flow rate. We altered the passage in line 262 (revised manuscript) to

…and V is the sample flow rate, here V = 0.283 std L min^{-1}.

Line 250: How many OPC intervals were used, and are they have the same length in terms of time? MDC = 1 count is defined. How this is assumed or calculated?

Each OPC interval is 5 sec in duration (can be set in the OPC). A typical background measurement is 5 minutes, thus 60 intervals. For every INP measurement, there is one background measurement before and after, so 120 intervals in total. The INP measurement is typically 15 min in duration, which is equivalent to 180 intervals.

The minimum detectable concentration is the smallest non-zero signal from the OPC, which is 1 count, divided by the volume of air sampled during the INP measurement, which is 15 min * 0.283 std L min^{-1} = 4.245 std L. Therefore, for the defined INP sampling conditions the MDC is 1/4.245 = 0.2356 INP std L^{-1}. To reduce the level of ambiguity we changed line 270 (revised manuscript) as follows:

The minimum detectable concentration (MDC) is 1 count (particle) in the ≥ 4 µm OPC size bin over a 15-minute INP measurement with a sample flow rate of 0.283 std L min^{-1} ~~over 15 minutes~~, which equals MDC = 0.236 std L^{-1}.

Line 269: clarify '. . .size bin 4995. . .'

According to the manufacturer, the used OPC (MetOne GT-526S) can count 4995 particles per second, and simultaneously classify their optical size and place them in one of 6 user-defined size bins. We changed line 298 (revised manuscript) accordingly:

According to the manufacturer, the used OPC can count 4995 particles per second, and simultaneously classify their optical size and place them in one of 6 user-defined size bins, with an overall accuracy of ± 10% to the calibrated aerosol.

Line 313: Is AF is same FF?

Yes, AF and FF are the same. We changed Figure X (revised manuscript) correspondingly:

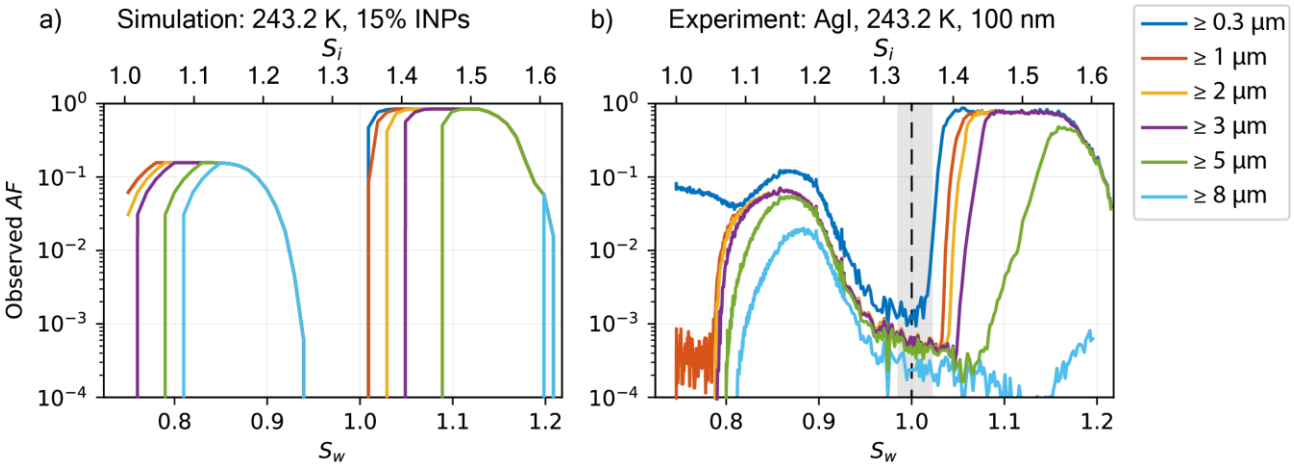

Figure 9. … with a prescribed fraction of INPs of 15% for S_w < 1 and b) … (See continuation of text after the next **reviewer comment**)

Furthermore, we changed line 351 (revised manuscript) correspondingly:

For the simulation, the fraction of INPs has been set to 15% for the simulation to agree best with the experiment. The fraction of INPs depends on the fraction of ice active ϐ-AgI particles within all particles

(Marcolli et al., 2016), which also contain ice inactive α-AgI particles and cannot be deduced by the 2D diffusion model, and therefore, needs to be prescribed.

Figure 9: Add vertical line Sw = 1.04 to panel b to understand AF value. Please comment on AF. Do you achieve maximum droplet activation?

We thank the reviewer for the comment. The aim of Figure 9 is to compare a measurement, where sedimentation of hydrometeors are observed, with the output of the 2D diffusion model. Therefore, we argue a comparison with the supersaturation used for INP measurements (S_w = 1.04) is unrelated to the shown experiment in Figure 9. Instead we add a dotted line to S_w = 1.0 where we also show the chamber uncertainty, and where the AgI particles activate to hydrometeors. We have now clarified the comments of the AF in the figure caption as shown below:

Figure 9. a) Simulated activated fraction curve as a function of S_w with a prescribed fraction of INPs of 15% for S_w < 1 and b) measured activated fraction curve of d_m = 100 nm silver iodide (AgI) particles, both at T = 243 K, with a particle residence time of τ = 13.7 sec at p = 965 hPa. Sizes stated in the legend indicate what fraction of all particles entering the chamber are activated and grow to or beyond the indicated size. Grey shading refers to chamber uncertainty around S_w = 1.0 (see Section 3.1 for details).

We added a sentence discussing whether maximum droplet activation is achieved in line 368 (revised manuscript).

The AF in the experiment as well as in the simulation is leveling off at AF = 0.85 for 1.02 ≤ Sw ≤ 1.13. Ice crystals (≈ 15 % in the experiment) and supercooled droplets are continuously formed at these supersaturations, but ice crystals grow to such large sizes that they sediment and are not detected at the outlet anymore. Therefore, maximum droplet activation of all non-ice active particles is observed in this region. The sedimentation of droplets is observed delayed at Sw ≥ 1.16 compared the model output at Sw ≥ 1.13. This is likely a result of the delayed onset of liquid droplet formation seen in the experiment. While increasing the supersaturation the droplets remain too small, thus their size-dependent settling is delayed, too

Figure 10: The tail end of the size distribution is not shown. It looks significant number of large particles exist. How these large particles (> 1 um) are distinguished from ice crystals?

During the rewetting procedures, when HINC-Auto is held at 25 °C, the chamber is sampling unfiltered air from the sampling line over a period of 2 minutes. During this period, only particles ≤ 2 µm where observed in the corresponding OPC size bins. We therefore distinguish these large particles (> 1 µm) by only using counts in the ≥ 4 µm OPC size to deduce the ice crystal number concentration. As shown in section 3.2.3 if active as CCN these particles are expected to grow to a maximum diameter below 4 µm ($d_{liq\ 2.0}$ = 3.31 at 965 hPa and τ = 9.1 s).

Section 3.3: It is not very clear how AF/FF values are converted to std L min-1 as shown in Figure 11. Please show the equation. Do you use std Temperature and Pressure values?

The AF/FF values are not converted to INP concentration in std L min^{-1}, instead the counts from the OPC are converted to INP concentration. All concentrations are shown in std L^{-1}, as pointed out by the y-axis label. As the flow rate with the mass flow controllers in the HINC-Auto system is set to std L min^{-1}, all concentrations (INP, LOD, MDC) are in std L^{-1} after using this flow rate in the calculations. The MFC measures the pressure and temperature and converts it to standard conditions. As the derivation of the INP concentration is discussed in section 2.4, we added an equation there to line 257 (revised manuscript) to show the derivation of the INP concentration. We moved ΣBG counts, $\Sigma N_{BG\ samples}$, V and t_{OPC} from the after the calculation of the LOD (eq. 2) to this section, as here they are used for the first time.

The INP concentration is calculated as follows:

$$INP\ concentration = \left(\frac{\sum INP\ counts}{\sum N_{INP\ samples}} - \frac{\sum BG\ counts}{\sum N_{BG\ samples}} \right) \frac{1}{Vt_{OPC}} \quad (1)$$

where ΣINP counts is the sum of all counts (particle number) in the ≥ 4 µm OPC size bin during the INP measurement, $\Sigma N_{INP\ samples}$ is the total number of OPC intervals during the INP measurement, ΣBG counts is the sum of the background counts (particle number) in the ≥ 4 µm OPC size bin before and after the INP measurement, $\Sigma N_{BG\ samples}$ is the total number of background OPC intervals before and after the INP measurement, t_{OPC} is the duration of each OPC interval in minutes (here 5 sec, thus 5/60 min), and V is the sample flow rate, here V = 0.283 std L min^{-1}. As the volume flow through the OPC is controlled by the MFC in std L min^{-1}, the resulting INP concentration is INP std L^{-1}.

It is not clear here, but how data is quality controlled? How data is flagged as good or bad. Any outliers are removed? Thoughts on data quality assessment would useful.

This is a very valuable comment, for which we would like to thank the reviewer. We added a corresponding section 2.5, line 272ff (revised manuscript):

2.5 Quality control

The algorithm to derive the INP concentration also performs a quality control. When a deviation from the set conditions (see below) is observed, the data is stored normally but a flag is added to the measurement. The evaluation of the flag and a potential exclusion of the data needs to be done by a researcher during post processing. Deviations are flagged (i) if the mean temperature of either wall is off by more than a predefined value (here ±0.15 K), (ii) one of the two MFCs reports a deviation between the set and the measured flow rate by more than ±50 std mL min^{-1}, (iii) the chiller reports an error, (iv) the pressure within the chamber is different by more than 50 hPa from the ambient pressure or (v) the water reservoir, used to rewet the chamber walls, is below a defined threshold (~100 mL).

References

Murphy, D. M. and Koop, T.: Review of the vapour pressures of ice and supercooled water for atmospheric applications, Quarterly Journal of the Royal Meteorological Society, 131, 1539–1565, https://doi.org/https://doi.org/10.1256/qj.04.94, 2005.

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