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Interactive comment on "Novel estimation of aerosol processes with particle size distribution measurements: a case study with TOMAS algorithm" *by* Dana L. McGuffin et al.

Anonymous Referee #2

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In this work, the authors apply an inverse modelling method based on inventory control to an aerosol box model. The application of this method to aerosol nucleation and growth is novel and interesting, and I look forward to seeing what insights can be gained from its application in a more comprehensive modelling study. The paper is well-written, and merits publication provided that my following comments are addressed:

General comments:

In reality, emissions (and other processes) vary on a wide range of time scales, including seasonal, weekly, and diurnal cycles, and variability at shorter timescales. These variations are likely not fully captured by any CTM. Does this present a challenge for

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Discussion paper

the choice of an appropriate value for the gain K_c? Can the authors comment on the robustness of their method to this issue?

Could the authors discuss the sensitivity of the results to the sink rate chosen in this work? I understand that a detailed treatment of dilution, transport, and deposition is beyond the scope of this study. However, given that the rates of these processes are both variable and uncertain, I think that a little further discussion is warranted. This discussion would also inform the potential of this method to be applied in a CTM, as there are uncertainties in more model processes than can be tested simultaneously using this method.

Technical comments:

I do not see the chosen values of the gain K_c listed in the paper. It would be best to list them in section 2.2. It may be helpful to express them as convergence timescales.

p4, line 20: Is it improved performance that the authors anticipate, or greater understanding? I would guess that the authors would find similar or better performance in using the mixing ratios directly as the control variables.

p8, line 10: What is the timescale of the moving average? The authors later state that the synthetic noisy measurements are filtered with an 11-hour timescale. Was the same timescale applied to the observations?

p9, line 1, "will repeat": The authors should use the present tense here. Reserve the future tense for future work.

p10, line 31-33. I found this sentence confusing. If I am reading Fig. 7a correctly, the maximum in normalised mean bias increases from 0.06 to 0.09. Is the "maximum bias" the authors are referring to, then, the bias for a single time step of the box model (not shown in the figures)? If so, I would request that this be split into two sentences, as the second half does not refer to Fig. 7a.

p5, line 1 states that the TOMAS model simulates particles as small as 0.5 nm in di-

ameter. However, p12, line 31 seems to indicate that nucleated particles are generated with an initial diameter of 3 nm. Are the smallest size bins unused (i.e. always contain zero mass) in this study? This should be stated plainly in the methods section.

p13, lines 9-10: It may be clearer to say “this is after sunset on February 22 and before sunset on March 28th”.

Figure 7: Does the mean bias in the nucleation rate decrease when noise is added to the synthetic measurements? If so, this warrants a brief discussion in the text.

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