The novel methodological aspects of the work are described in Chapter 2 of the thesis. I find this chapter somewhat confusing, because it appears to have three distinct purposes. Firstly, the chapter provides some general introductory material, which has substantial overlap with the introductory chapter. Secondly, the work done by the candidate to recalculate the OMI HCHO column measurements is described. These revised data are used in the subsequent chapter on isoprene inverse modelling (and presumably also by Lieschke et all, 2019). Thirdly, several filtering techniques are described, which it seems are only relevant for the isoprene inverse modelling work. I do understand the logic of including all of this material in a chapter called “Data and Modelling”, but I think a separation into distinct chapters would help the reader, and also make it clearer what the candidate has contributed. In particular, for the “middle part” of this text, it would be good to see a section similar to Section 4.9, in which the contributions of different workers (including the candidate) are clearly described. Subsequent work published (and even in preparation) using the new HCHO dataset could also be mentioned here, which would help to show how useful this work has been for colleagues and the wider community. Perhaps the work on filtering the HCHO columns (Section 2.7) could be merged into Chapter 3, especially if this will all be submitted as a standalone paper anyway.

My main criticism of Chapter 3 is that the work is not described adequately in the context of previous studies. A top-down study by Bauwens et al. (2016) is mentioned in passing in Section 3.1.2, but almost no details are given. This study appears to be highly relevant, given that it seems to use an almost identical method. Chapter 3 must include a comparative discussion of the method used in this thesis and the method of Bauwens et al. (2016), and how the results of the two studies are influenced by their choice of methodology. Similarly, the range of values for Australian isoprene emissions (both bottom-up and top-down) given in Table 3.1 should be discussed in more detail.What is it about the different emission models and inverse approaches that lead to such a large range of values? Can the results of this work be understood simply in terms of the way the methodologies are applied, or has something genuinely new been learnt? In praise of Chapter 3, I really appreciated the detailed examination of uncertainties in the analysis, and in particular the thorough discussion of the role of the isoprene-HCHO yield and its implications for future work.

I noticed several small apparent errors of understanding in Chapter 1 which the candidate should revisit while revising their thesis. For example, on page 3, the hydroperoxyl radical (HO 2 ) is referred to as “hydrogen dioxide”, a name which is never used in the relevant atmospheric chemistry literature. I was also not convinced by some aspects of the candidate’s description of ozone chemistry. For example the photolysis reaction of ozone in Equation Set 1.2 (page 4) is claimed to proceed at wavelengths less than 1180 nm. Does the candidate have a reference describing how quickly the reaction proceeds at such low photon energies? The absorption cross section for ozone photolysis used in the MCM (Master Chemical Mechanism) is only defined below 350 nm, with wavelength-dependent quantum yields for O( 1 D) and O( 3 P) below 350 nm (http://mcm.leeds.ac.uk/MCMv3.3.1/parameters/photolysis.htt). Significant amounts of ozone photolysis at lower photon energies would result in a much shorter lifetime for tropospheric ozone than predicted by the MCM (or any other model which I have used). Furthermore, the high abundance of “M” at lower altitudes is given as a reason for the existence of the ozone-layer, but M are actually required to stabilise ozone produced by photolysis in the stratosphere (Equation Set 1.2), where they are less abundant. The candidate should rethink and revise their text here.

I also think that the candidate should give a better overview of the different kinds of sources of VOC in Section 1.3. The candidate primarily focuses on biogenic isoprene, which is reasonable given the focus of the thesis, but anthropogenic and pyrogenic VOC are barely mentioned, and not until later in the text. Also, “reactivity” is listed as one of the fundamental properties of VOC near the beginning of Section 1.3, but I miss some discussion about what determines the reactivity of different VOC, and what this means for their atmospheric lifetimes. This is especially important, since the atmospheric lifetimes of both isoprene and HCHO are important concepts which are drawn upon in several places later in the thesis.

Given their importance in the thesis, I would also like to see more understanding of biogenic isoprene emissions demonstrated by the candidate in Chapter 1, especially how they are modelled, and the basis (or lack thereof) that this emissions modelling has in actual measured data. BVOC emission models, specifically MEGAN, are mentioned in several places in the thesis. In many of these cases literature references are given, and particular aspects of the potential shortcomings in these models are described, such as a poor representation of soil moisture, or an over-reliance on measurements from younger trees. Instead of scattering this information throughout the thesis it should be collected in one place where the candidate can demonstrate an understanding of how these models work and what their known shortcomings are. The results in subsequent chapters can then be related back to the material from the introduction

As a matter of style, tables and figures should not be presented in the conclusions chapter. Figure 5.2 is particularly superfluous, since it is exactly the same as Figure 3.7