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Comment

***Interactive comment on “A Lagrangian model of air-mass photochemistry and mixing using a trajectory ensemble: the Cambridge Tropospheric Trajectory model of Chemistry And Transport (CiTTYCAT) version 4.2” by T. A. M. Pugh et al.***

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We thank Dr Taraborrelli and the anonymous reviewer for their helpful and constructive comments, which we address in turn below.

**RC:** The limonene oxidation mechanism by Stockwell et al. (1997) here used artificially produces methacrolein (MACR) that is only an isoprene product. This is a source of error especially when the model is used:

- to reproduce the MVK/MACR and (MVK+MACR)/ISOP ratios

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- to constrain isoprene emissions

- to estimate HO<sub>x</sub> constraining by the photochemical age as in Karl et al.(2007)

This source of error likely explains part of the model overestimate for MVK+MACR reported by Pugh et al. (2010b). Therefore, this problem should be acknowledged and an alternative should be either planned or implemented.

**AC:** We agree that the limonene oxidation mechanism of Stockwell et al. (1997) is non-optimal. However, this was (and to our knowledge remains), the only available limonene oxidation mechanism at this intermediate level of complexity.

The contribution of MACR from the limonene scheme to the overall MACR+MVK reported in Pugh et al. (2010) is small (9%). This does not change the conclusions of Pugh et al. (2010). Indeed the majority of the model overestimate is attributable to MVK, which is not formed by the limonene scheme. We agree, however, that this is a limitation of the current version of the model, and we will add the following to pg. 2481 line 17 of the revised manuscript:

'This limonene scheme should be used with caution in studies where the isoprene-exclusive oxidation product methacrolein (MACR) is a key output, as it apportions a fraction of limonene oxidation products to MACR.'

**RC:** Regarding soil NO<sub>x</sub> emissions there is a recent algorithm (Steinkamp and Lawrence, 2011) that builds on Yienger and Levy (1995) and has been evaluated against measurements. This algorithm could be mentioned and its inclusion planned.

**AC:** We have modified the revised manuscript to refer to this new addition to the literature on pg. 2484, line 8:

'Other databases exist, however, the global emissions of soil NO<sub>x</sub> inventories show significant variation from 5.5 Tg N yr<sup>-1</sup> (Yienger and Levy, 1995), through 8.6 Tg N yr<sup>-1</sup> (Steinkamp and Lawrence, 2011) and 9.7 Tg N yr<sup>-1</sup> (Potter et al., 1996) to 21.0 Tg N yr<sup>-1</sup> (Davidson and Kinglerlee, 1997) and a decision between them cannot currently be

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made on the basis of measurement validation.'

We do not currently envision updating the soil NO<sub>x</sub> dataset in CiTTyCAT in the near future. However, it would be straightforward for a user to implement the Steinkamp and Lawrence algorithm.

**RC:** A number of other models based on Lagrangian transport have been developed and used over the years. For example NAME (e.g., Rigby et al., 2011), Attila (e.g., Stenke et al., 2007), Atlas (e.g., Wohltmann and Rex, 2009), CIAMS (e.g., Konopka et al., 2010), Lagranto (e.g., Wernli and Davies, 1997; Cui et al., 2009), and Flexpart (e.g., Stohl et al., 2005; Cui et al., 2009). These models differ in design and are constructed for different research issues. However, there is some overlap of the CiTTyCAT model with these other Lagrangian models. Therefore I suggest including a brief discussion in the paper of other Lagrangian methodologies and how (and perhaps why) the approach adopted for CiTTyCAT model differs from the other approaches.

**AC:** The following discussion of Lagrangian models and their relation to CiTTyCAT is now given before the last paragraph of Section 1.

'CiTTyCAT calculates the evolution of air mass composition, along one or many given trajectories. The trajectories are calculated by a separate model. In most applications of CiTTyCAT meteorological analyses from the ECMWF are used to calculate trajectories using either the ROTRAJ model (Methven, 1997) or FLEXTRA (Stohl et al, 1995). Several other Lagrangian models take a different approach, and calculate both trajectories and composition, in a variety of different ways. For example, ATLAS (Alfred Wegener Institute Lagrangian Chemistry/Transport System, Wohltmann and Rex, 2009) calculates trajectories throughout the stratosphere (with a basic troposphere), and optionally includes a chemical scheme with 49 tracers and 170 reactions. ATLAS was designed to be used for both high-resolution process studies and for decadal simulations of the global stratosphere, and handles both transport and chemistry in one model.

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An extension of trajectory models (which use the resolved winds to calculate trajectories) is the Lagrangian particle dispersion model (LPDM). LPDMs release particles that are advected by large-scale winds plus a stochastic term to represent unresolved turbulent motions. A well-used and cited LPDM is FLEXPART (Stohl et al, 2005). The UK Meteorological Office's NAME model is an LPDM that now includes the STOCHEM chemistry scheme (70 chemical species and 160 gas-phase reactions, Collins et al 2000). NAME uses a hybrid Lagrangian-Eulerian approach: the chemistry is calculated on a fixed grid, where all particles located within a grid cell are brought together for the chemistry calculation (Jones, 2007).

Using either approach (trajectory or LPDM) it is necessary to exchange material between trajectories to represent mixing. In LPDMs turbulent mixing is represented by the random walk of particles and then a grid is imposed and the properties of particles within grid cells are mixed by averaging. With passive tracers this can be done once at the end of the trajectories (a retroplume calculation). There are choices to be made about the grid-size and degree of mixing. However, to represent the nonlinear coupling between mixing and chemistry it would be necessary to perform regular re-gridding. CiTTYCAT takes an alternative approach to mixing. Rather than use a random walk, it is assumed that mixing is dominated by diffusion of a vertical profile (obtained by averaging the trajectory ensemble) plus a constant flux parameterisation in the boundary layer. The advantage of this approach is that the mixing ratio tendencies associated with mixing can be added to those from photochemical reactions so that the processes are coupled on the ODE integrator time-step (rather than a much longer re-gridding timescale). This is particularly important for short-lived species such as NO<sub>x</sub> and isoprene.

As CiTTYCAT uses pre-calculated trajectories, many sensitivity tests can be carried out on the same set of trajectories. Using a LPDM, this is not possible, as the stochastic parameterisation of turbulence means that the same particle releases will not follow exactly the same paths each time they are run.

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The Edinburgh – Lancaster Model for Ozone (ELMO-2) is a UK-specific Lagrangian CTM (Strong et al, 2010), which simulates chemistry along HYSPLIT trajectories (Draxler and Rolph, 2003), pre-calculated backwards from a PBL measurement. ELMO-2 includes the STOCHEM chemistry scheme and a well mixed PBL, however there is no mixing when trajectories are in the free troposphere. Although ELMO-2 and CiTTyCAT are based on the same approach, the latter is more sophisticated in its treatment of chemistry and mixing.’

**RC:** The vertical coordinate of the model seems to be pressure surfaces (although this is not always clear, see below). I suggest some discussion why this choice was made compared to a variety of other possible choices (Kasahara, 1974).

Also

**RC:** p. 2502, l. 1: Here it seems that the vertical coordinate is a hybrid-pressure coordinate, earlier in the paper pressure is mentioned as the vertical coordinate. It should be clearly stated here which vertical coordinate (and thus which vertical velocity) is used to calculate the trajectories.

And

**RC:** p. 2476, l. 11: this is no minor point. There is a variety of vertical coordinates that could be envisaged (e.g., Kasahara, 1974). I suggest more discussion here.

**AC:** Since the model is Lagrangian, the vertical coordinate is simply used to label position of air parcels relative to other fields. However, the fields used in the parameterisations are typically discretised in terms of values on vertical levels. The vertical discretisation differs between routines and so does the vertical coordinate.

The ROTRAJ model (as the UGAMP trajectory model will henceforth be called) uses analyses on the ECMWF model (hybrid-pressure) levels to calculate the trajectories. Their vertical locations are output both in terms of a pressure and height value (related through hydrostatic balance in the ECMWF model).

The CiTTyCAT mixing scheme and treatment of the boundary layer (Sections 2.1 and 4.2.1) are formulated using height to define the background profiles, boundary layer top and vertical diffusion. Boundary layer height is obtained from the ECMWF driving model at the location of each trajectory.

Some of the schemes, such as photolysis and its associated ozone climatology, use pressure as the vertical coordinate since this is the form of the input data.

**RC:** Finally I suggest bringing clearer across for which time scales (in my view several days but less than say a month) the model is designed. This is especially important in the abstract, conclusions and introduction.

**AC:** The relevant timescale was mentioned in Section 1. It ranges between the average stretching rate (1-2 days) and the free tropospheric mixing timescale (7-15 days). The actual length of useful integration is rather case specific and depends on the trajectory ensemble behaviour, mixing rates and contrast in composition with neighbouring air masses.

**RC:** p. 2476: I am a bit confused here: going from ‘Eulerian’ to ‘Lagrangian’ means changing the frame of reference so it is more than just manipulating equations. Also: Is Eq. 2 valid for compressible flow? I suggest a bit more discussion here.

**AC:** There are no approximations made and the steps from (1) to (2) are exact. The continuity equation and definition of  $D/Dt$  is added to make this step clearer. Pg. 2476, line 19 now reads:

‘Combining with the mass continuity equation:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \underline{u}) = 0 \quad (1)$$

yields the classical Lagrangian equation for mixing ratio:

$$\frac{Dc}{Dt} = S - \frac{1}{\rho} \nabla \cdot \underline{F} \quad (2)$$

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where the Lagrangian derivative  $Dc/Dt = \partial c/\partial t + \underline{u} \cdot \nabla c$  represents the rate of change following fluid parcels.'

**RC:** p. 2477, eq. 3: is  $h$  a constant? If it is not state on which variables it depends on.

**AC:**  $h$  is prescribed on the input trajectory, or in absence of this, by a internal dataset of boundary layer height (Law and Pyle, 1993, we emphasise that this dataset is for first estimates only), as described in Section 3.1. We have changed pg 2477, line 6 to read, '...across the boundary layer of prescribed depth,  $h$  (cm).'

**RC:** p. 2477: It is not obvious from the discussion why dry deposition is dependent on  $h$  whereas  $r_{wet}$  is not. I suggest more discussion here.

**AC:** Wet and dry deposition are described in Section 2.5. We have therefore added a rider on pg. 2477, line 22 reading '(see Section 2.5 for details of the deposition schemes)'. Further, on pg. 2489, line 24, we have added a sentence reading: 'Dry deposition velocity,  $V_{dry}$ , determines the dry deposition flux of a species at the surface according to the concentration of that species at a reference height (usually 10 m or less). In CiTTYCAT, species within the boundary layer are assumed to be well mixed, and thus the surface dry deposition flux is apportioned uniformly across the depth of the boundary layer via  $V_{dry}/h$ .'

Wet deposition rates are calculated in a different manner to those for dry deposition (Section 2.5.2) and are independent of boundary layer height,  $h$ . In the driving ECMWF model the vertical distribution of precipitation is unknown and the precipitation is assumed to reach the ground instantaneously upon condensation from any level in the column above. Therefore, wet deposition (in precipitation) is considered to take constituents from any level (unless descending), while dry deposition requires turbulent motions to bring air in contact with the ground and is therefore only acts on boundary layer air.

**RC:** p. 2478., l. 21: This is very hard to understand without having read the following

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sections of the paper. Suggest to rephrase.

**AC:** We have rephrased this section to read: 'In ensemble trajectory mode, the diffusive flux divergence term is modelled by mixing between multiple (usually more than 100) trajectories, each of which evolves photochemically. In this case, vertical mixing between the trajectories propagates the effect of surface fluxes beyond the boundary layer top.'

**RC:** p. 2479: Chemistry schemes: there should be some discussion here of heterogeneous chemistry. It is neglected but is this assumption justified?

**AC:** Heterogeneous chemistry is not included in the current version of CiTTYCAT, as it has not been relevant for the recent uses of the model (trans-Atlantic transport and gas-phase VOC chemistry). Heterogeneous reactions have been included in the past however (Wild, 1995), and the facility to incorporate them using DELOAD remains. A short section entitled '2.2.5 Heterogeneous chemistry' has been added, which reads:

'This version of CiTTYCAT does not include heterogeneous chemistry, although DELOAD is able to handle such reactions. Including such chemistry requires assumptions regarding aerosol/cloud particle composition and size which have not been justifiable in recent applications of the model. Quasi-heterogeneous chemistry is included in the gas-phase for  $N_2O_5$  hydrolysis. Future applications of the model must assess whether heterogeneous chemistry is important for their case, and add reactions if necessary.'

**RC:** p. 2479., sec 2.2.1: I do not understand: If I want to change chemical rate constants I think I should rerun DELOAD but in l. 18/19 the opposite statement is made.

**AC:** DELOAD formats the production and loss terms for the ordinary differential equations, but the rate coefficients may be changed independently of DELOAD. We have slightly modified the second sentence of Section 2.2.1 to avoid this confusion. It now reads: 'The CiTTYCAT reaction files for bimolecular, termolecular and photolysis reactions are stored as ASCII files that can be fed to the DELOAD programme (Nejad, 1986;

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Brown et al., 1993), which then generates Fortran code containing the production and loss terms for the ordinary differential equations.'

**RC:** p.2480, l. 19: Perhaps it is worth mentioning also other studies suggesting problems in our understanding of organic compounds emitted by plants (e.g., Kiendler-Scharr et al., 2009).

**AC:** This sentence now reads: 'Given the current uncertainty in isoprene chemistry and its importance for gas- and aerosol-phase chemistry (e.g. Lelieveld et al., 2008; Kiendler-Scharr et al., 2009; Pugh et al., 2010b; Stone et al., 2011), the absence of tuning of reaction rate coefficients in MIM2 (in order to improve the fit for certain species at the expense of others; a common practice in condensed mechanism development), is useful as modifications can be made to the chemistry, without unintentionally detuning the scheme.'

**RC:** p. 2481, l. 2: citation for the photolysis scheme.

**AC:** We have added a reference to Section 2.3 here.

**RC:** p. 2481. l.18: why should the monoterpene schemes not impact say the OH chemistry of the model?

**AC:** The monoterpene schemes do impact the OH chemistry of the model when monoterpene mixing ratios are greater than zero. These schemes provide a pathway from the initial monoterpene, through monoterpene-specific intermediate species, to intermediate species which are common with other schemes (e.g. CH<sub>3</sub>OO). As the reactions of these monoterpenes are only favoured in the forward direction, if no monoterpenes are contained within the model then these schemes are unused and do not influence the model result.

**RC:** p. 2482, l. 3: state on which measurements the ozone climatology is based on. Furthermore, I suggest adding the dataset as an electronic supplement so that others can also use the climatology.

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**AC:** The first two sentences of this paragraph now read: ‘As the O<sub>3</sub> column shows considerable latitudinal variation, a zonally averaged O<sub>3</sub> climatology (Li and Shine, 1995) is read in according to the model latitude and month. This climatology used the Solar Mesosphere Explorer near-infrared airglow data and is available at [http://badc.nerc.ac.uk/data/ugamp-o3-climatology/ugamp\\_help.html](http://badc.nerc.ac.uk/data/ugamp-o3-climatology/ugamp_help.html).’ We have also added to the end of this paragraph: ‘Alternatively, the US Standard Atmosphere can be used.’

**RC:** p. 2482, l. 19: How realistic is the assumption of 0.1? why is the optical depth not allowed to vary?

**AC:** The global mean aerosol optical depth (at 550 nm) is close to 0.1. Reddy et al. (2005) and Chin et al. (2002) suggest 0.12 and 0.135 respectively. The optical depth can easily be varied if needed, but this value is used as a default so that the effects of aerosol aren’t overlooked. We have rephrased this sentence to: ‘A global mean column optical depth of 0.1 is used as a default (Hough, 1988).’

**RC:** p. 2482: I suggest giving a bit more detail about the emissions. Are they specified as a mass flux (mass/time)? What is the time resolution (monthly, daily) of the emissions? If the emissions are part of the surface boundary condition a flux in mixing ratio is required – correct?

**AC:** This is correct. The emissions are specified as a number flux for each species in molecules  $\text{cm}^{-2} \text{s}^{-1}$ . We have modified the sentence beginning pg. 2483, line 10 to reflect this: ‘Emission fluxes are read in at each model step, converted to number fluxes (molecules  $\text{cm}^{-2} \text{s}^{-1}$ ), and instantaneously mixed over the prescribed boundary layer height (if simple or no mixing is used).’ The time resolution varies with the dataset and we have checked that this information is included in the individual descriptions.

**RC:** p. 2488, l. 20 change ‘diverges’ to ‘differs’.

**AC:** Done

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**RC:** p. 2490, l. 28: can a model be ‘informed’?

**AC:** We have changed this to ‘parameterised.’

**RC:** p. 2491, l. 6: what is meant by ‘correction to the units’?

**AC:** This refers to a coding error in a previous version of CiTTYCAT. On reflection, we feel this is an unhelpful detail in this paper and have removed it. We have added the following relating to code maintenance to Appendix A: ‘Model documentation beyond the current paper exists primarily as in-code comment; further descriptions of algorithms and rationale for choices are provided in Wild (1995), Evans (1999) and Cain (2009). Release of a new model version is by consensus of the authors of this paper. Error-trapping is facilitated by code-sharing and identified errors are logged and corrected within the code.’

**RC:** p. 2492, l. 23: Mention the numerical method used in DVODE. Below the term ‘Jacobian Method’ is used but no explanation or reference is given.

**AC:** This sentence now reads: ‘Coupled ordinary differential equations for the evolution of mixing ratios (Eq. 5) are integrated using the DVODE (Variable-coefficient Ordinary Differential Equation) solver using variable-coefficient Adams-Moulton and Backward Differentiation Formula methods in Nordsieck form with a variable time step (Brown et al., 1989).’

**RC:** p. 2494: Box mode: I know this is a difficult question but what is the size of the box that is assumed here. Possibly, depending on the issue in question the size of the box may vary?

**AC:** The size of the box will be defined by the case in question and the way a user applies the model. In the free troposphere the box may be considered as a point, or deemed to represent an area with a characteristic length scale of 100 km or more. For boundary layer studies the instantaneous boundary layer mixing assumption sets an effective lower limit of  $\sim 5$  km for the characteristic length (see pg. 2487, line 2). As box

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size is so dependent on how the user applies the model, we do not feel it appropriate to set specifications (other than that just described).

**RC:** p. 2492, l. 6: ‘the model is in the boundary layer’?

**AC:** Unlike the dry deposition scheme, wet deposition is not limited to the boundary layer. See discussion of wet and dry deposition above. We have rephrased this to read, ‘the air parcel is in the boundary layer’.

**RC:** p. 2497, l. 27: You mean ‘ozone concentrations’ correct?

**AC:** Yes, we have now made this explicit. For consistency, we have also replaced ‘concentrations’ with ‘mixing ratio’ throughout the paper where the word is used to refer to the model variables, inputs or outputs.

**RC:** p. 2499, l. 26: state for how long typically the trajectory is considered, 10 days? Or longer?

**AC:** We have added ‘5-day’ before ‘trajectories’ on pg. 2500, line 1.

**RC:** I suggest somewhat more discussion here on the diffusivity  $\kappa$ . How well is this parameter known. Would a different value be deduced, if a different set of species would be used? How sensitive are the results of the model simulation to variations of  $\kappa$ ?

**AC:** We have added the following the the final paragraph of Section 4.2.1: ‘In other cases,  $\kappa$  may not be so well constrained by observations, and must be estimated, dependent on the meteorological situation. Under stable conditions,  $\kappa \simeq 0.5 \text{ m}^2 \text{ s}^{-1}$  would be typical. If the meteorological conditions are more turbulent,  $1.5 \text{ m}^2 \text{ s}^{-1}$  would be more suitable. The authors suggest that  $\kappa$  is estimated based on observations if possible, or a comparison with literature values, and that sensitivity tests are carried out to establish the uncertainty in this parameter. In cases where there are weak gradients in the background profile, varying  $\kappa$  may only have a small effect on the results. However, if there are strong gradients (e.g. a biomass burning plume), the

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choice of  $\kappa$  may be important.’

**RC:** p. 2505: There should be more discussion here on the issue of ‘too much divergence’. Can you quantify what is ‘too much’? Further, there could be interesting atmospheric situations where trajectories diverge, so isn’t it a shortcoming of the model that such situations need to be excluded? Could there be a way to overcome this shortcoming.

**AC:** We have added the following after ‘diverge too much.’ on pg. 2505, line 5. ‘This is a judgement that must be made based on the case in question. A column of evolving background trajectories follows each trajectory separately. Every mixing time step (typically 1 hour), the background profiles are averaged. In some cases, the trajectory ensemble may diverge such that this averaging is inappropriate. For example, if a plume splits with one half passing over ocean and the other half passing over a continent, then it may be better to simulate the two sub-plumes separately.’

**RC:** p. 2508, l. 9: give a reference for Fast-J.

**AC:** Done.

**RC:** Fig. 10: I cannot find the orange arrow.

**AC:** The arrow is located on the y-axis. We have modified the caption to make this more obvious.

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