

John M. C. Plane, Jörg Gumbel, Konstantinos S. Kalogerakis, Daniel R. Marsh, and Christian von Savigny

Response to the Editor's comments

We thank the editor for a very careful reading of the manuscript and for making a number of helpful suggestions for improving it. The editor's comments are italicized, and our response is in normal typeface. In the revised manuscript with marked changes, the changes are shown using track changes.

I have some comments of my own. See below. Please consider them and make revisions as you think appropriate. You do not need to provide full responses (but of course responses are welcome and might be interesting).

One referee suggested modifying the title and you argued against that. I guess that you and your co-authors are a bit worried that mesospheric experts who are not chemists will criticise this because it doesn't cover their speciality in sufficient detail. My own comment having read the paper is that there is plenty in it that is interesting to me, not a mesospheric expert, and it would be a pity if the paper was ignored by a broader readership because of the 'Chemistry of' in the title. The 'Opinion' in the title is a useful indication that this not claiming to be complete and authoritative. I would be in favour of dropping 'Chemistry of the'. The abstract could say 'The emphasis here is on chemistry, but we also discuss ...'. But it is your call.

Author response: this is a very fair point. We have therefore removed “Chemistry” from the title, and changed the Abstract as suggested.

Detailed comments:

40: 'pressures falling from 0.1 mbar to below 1 mubar, at which point molecular diffusion becomes more important than eddy diffusion in transporting constituent species' -- I think that you are saying that where does molecular diffusion become more important at about 1 mubar? But your previous characterisation of the MLT as up to 120km surely implies a much smaller pressure at the upper bound of that region?

Author response: this sentence now reads “The MLT is therefore subject to extremes: pressures falling from 0.1 mbar to below 0.1 μ bar (above the turbopause at a pressure around $\sim 0.5 \mu$ bar, molecular diffusion becomes more important than eddy diffusion in transporting constituent species); and temperatures ranging from below 100 K (the coldest part of the planet) to over 2500 K in ablating cosmic dust particles.”

Figure 1: Good that it is included. Some parts are a bit mysterious -- e.g. I suppose that the arrows above H₂O, CH₄ etc imply upward transport? There are many arrows on the diagram representing many different processes. I recommend that, having taken the trouble to provide the Figure, you provide a bit more information in the caption.

Author response: this is a good suggestion. The processes described by the different coloured

arrows are now identified individually in the figure caption.

65: *Why 'relatively' too dense to allow in situ measurement?*

Author response: “relatively” is now removed, and the sentence reads: “The atmospheric density in the MLT is too high for *in situ* measurements by satellites because of the resulting aerodynamic drag, and so satellite instruments rely on remote sensing ...”.

71: *'and by the vertical transport of atomic O' would be clearer.*

Author response: done.

79: *'impact of atmospheric tides in the MLT' -- you made a change in response to a referee comment. Their point was that it was in detecting the impact on composition of the tides that the emission observations had been useful. 'Impact of atmospheric tides on the MLT composition' would be closer to what the referee was recommending.*

Author response: done.

82: *'SNOE' -- defined only in acronym list -- giving definition when first used, as you have done for most acronyms, would be helpful.*

Author response: done.

93: *'contraction' -- also 'shrinkage' used elsewhere. Personally I don't like this kind of term because I think it can be confusing in the wider context. ('The mesosphere is shrinking!') Of course what you mean is that the physical criteria used to define the boundaries of the MLT, or boundaries within the MLT, do not correspond to fixed altitudes and changes in the temperature structure of the atmosphere therefore imply that these boundaries may change. A one-sentence explanation of this might be helpful.*

Author response: “Contraction” is a commonly used term in MLT science (e.g. in the title of the Mlynczak et al. (2022) paper we cite). We now define the term: “..., where contraction is defined as a decrease in the vertical distance between the same pressure levels”. “Shrinkage” has been changed to “contraction” throughout the paper for consistency.

98: *Is this a definition of 'sporadic E layers'?*

Author response: the definition is now better defined: “Sporadic *E* layers are ionospheric irregularities that consist of high concentrations of metallic ions and electrons in narrow layers”

138: *'A new metal' -- not really new, but newly observed in the mesosphere.*

Author response: agreed. This is now rephrased: “The metal Ni has been observed for the first time...”

149: *'metals often occur in pronounced layers' -- are these 'sporadic E layers'?*

Author response: no, these are neutral metal atom layers. This is now made clearer: “... these measurements have revealed that the neutral metal atoms often occur in pronounced layers, ...”

161: *'PMSE' -- give definition here as well as in acronym list.*

Author response: done.

183: *'energy of a photon absorbed in the thermosphere warms the mesosphere, thereby contributing*

to the transport of energy via thermal diffusion' -- but you seem to imply in the previous sentence that the downward diffusion of atomic oxygen is what transports the energy downward -- so isn't this 'via downward diffusion of atomic oxygen' rather than 'via thermal diffusion'?

Author response: we agree this is somewhat confusing. The sentence is now shortened to: “Thus, an odd oxygen (O or O₃) is converted back to O₂ and the energy of a photon absorbed in the thermosphere warms the mesosphere.”

194: 'The same circulation pattern ...' -- 'brings NO and CO downwards from the thermosphere' would be clearer -- but it was also be clearer to re-order the sentence, since you are in the previous sentence talking about the summer pole.

Author response: agreed. Now changed to “The same circulation pattern brings H₂O and CO₂ upwards over the summer pole, and NO and CO downwards from the thermosphere into the wintertime polar mesosphere at the opposite pole ...”.

212: 'polar summer mesopause' would be clearer.

Author response: done

214: 'NLC/PMCs' -- again better to give definition of PMC here. Is 'PMC' simply an alternative term for NLC?

Author response: PMCs are now defined at line 138 (in the revised manuscript).

217: 'observational data' rather than 'experimental data'? (This wasn't a lab experiment.)

Author response: done.

231: 'still eastward' a bit odd. 'Continuing eastward'?

Author response: done.

239: 'leading to an average period of close to 27 days' not needed?

Author response: this sentence has been made clearer: “The 27-day cycle is caused by the differential rotation of the Sun, with a period which is slightly variable but averages close to 27 days.”

280: '3.4 Energetic particle impacts' -- 'impact' is a word used many times in the article to mean 'effect' and we all have our favourite words. But here it gets genuinely confusing.

'Global observations of NO have been widely used to study particle impact ionization in the MLT region, e.g., showing a clear impact of particle forcing well into the mesosphere'

A reader might wonder whether the title of the section should be 'The impact of energetic particle impacts'?

Author response: we agree that “impact” is used ambiguously. “Impact” is now used exclusively to refer to collisional impacts (of meteoroids, energetic particles etc.), and “effects” to refer to the consequences of various processes. Section 3.4 is renamed “Effects of energetic particle precipitation”.

304: 'quasi-2-day-wave activity' -- is the 'quasi-2-day' important? i.e. is the reader supposed to interpret something particular from the use of that term?

Author response: we have added the following brief statement about this phenomenon “... (the quasi-2-day-wave is a planetary wave that is particularly significant in the mesosphere during summer)”

320: *'The recent study by Hoffman et al' -- clearer as 'As noted previously, the recent study by Hoffman et al'.*

Author response: done

326: *Re 'shrinking', see earlier comment. Are radio reflection heights thought to correspond to particular pressure levels. Or to some other physical variables?*

Author response: yes, it is the altitude of the pressure level that decreases when shrinking/contraction occurs (see earlier response).

330: *'A long-term cooling trend in the middle mesosphere appears to be well established at a rate of about 3 K decade⁻¹ around 70 km, with perhaps a weak negative trend in the mesopause region (Beig, 2011)' -- I was a bit confused by the final part of the sentence. Is the point that the trend is clear in the middle mesosphere but not clear at the mesopause?*

Author response: this sentence has been rephrased to make it clearer: "A long-term cooling trend in the middle mesosphere appears to be well established at a rate of about 3 K decade⁻¹ around 70 km; the trend in the mesopause region is much smaller, though possibly also negative ..."

522: *'The strong updraft connected to this circulation ...' -- personally I don't like the use of 'updraft' to describe this kind of large circulation because 'updraft' and 'downdraft' are terms that are often used in convection and I believe that the phenomenon occurring in the MLT is very different to convection. 'Strong upwelling' would be better.*

Author response: done.

562: *'in the dry middle atmosphere' -- omit 'dry'? A reader might think that this refers to some part of the middle atmosphere - the 'dry' part rather than the 'wet' part.*

Author response: done.

575: *I've expressed reservations about use of 'shrinking' -- but actually here it seems OK because some explanation is given of what 'shrinking' actually is.*

Author response: changed to "contraction" for consistency with earlier discussion.

662: *'courser-scale' > 'coarser-scale'*

Author response: done

664: *'The MLT has weather, just as in the troposphere, and its evolution in time is not solely determined by external forcing either from above or below. As with any chaotic system, small changes in initial conditions can lead to large changes in the final model states.' -- this is an interesting statement, but have is there any concrete evidence on this -- i.e. that processes within the MLT itself can lead to chaotic/unpredictable behaviour? The Richter et al (2022) paper doesn't seem to address this, but perhaps I have missed something.*

Author response: the Richter et al. (2022) reference should have appeared earlier in the paragraph, and has now been moved. Richter et al. (2022) discuss sub-seasonal forecasting, but not on a timescale of hours, so we have inserted "potentially" into the relevant sentence. A reference to substantiate the statement about chaos leading to large changes in final states has been added. This section now reads:

"Second, it will likely become common practice to run ensembles of model simulations to capture the spread of possible atmospheric states that comes from geophysical variance (Richter et al.,

2022). The MLT has weather, just as in the troposphere, and its evolution in time is not solely determined by external forcing either from above or below. As with any chaotic system, small changes in initial conditions can lead to large changes in the final model states (Liu et al., 2009). Such ensembles can potentially be used to make forecasts of the MLT on timescales from hours to weeks, benefitting forecasting for space and tropospheric weather as well as sectors that rely on GPS and radio communications.”