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## ***Interactive comment on “The emission factor of volatile isoprenoids: caveats, model algorithms, response shapes and scaling” by Ü. Niinemets et al.***

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I had considerable difficulty in reviewing this manuscript because I found it a strange hybrid between a straightforward literature review and an opinion piece. If practical, I would prefer that the two be clearly differentiated in the manuscript. Although the title suggests that the focus of this review is the so-called BVOC emission factor, in fact it is a much broader critique of the current methodologies used to predict BVOC emissions. My review, therefore, will also be a little strange. . . a straightforward review of their review followed by some of my own opinions on the subject.

In general, the authors point out, correctly and in considerable detail, that great ad-

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vances have been made since the first BVOC emission models were proposed in the early 90's, and they argue that this information needs to be incorporated into a new generation of models. As a review of advances in our understanding of the physical and biological controls over BVOC production/emission at the leaf scale, it is comprehensive, summarizing a great deal of research, much of which was carried out by one or more of the authors themselves. If the criteria for judging a review paper are: 1) is it comprehensive? - 2) is it an accurate summary of current knowledge? - 3) is it up to date? - and 4) is it necessary? - then this submission scores well on the first three points. I'm not convinced it was necessary, but I would certainly turn to it for a very useful summary, obviating the need to search back through the literature. In short, as a simple review of leaf-level controls of BVOC emissions, it stands up well and is a useful contribution.

Instead of being written as a straightforward review of controls over emissions, however, the authors have chosen to couch their review as a critique of the so-called 'base emission factor' or 'emission capacity' (Es) currently employed in all the emission models in common use. They argue that, in light of the considerable advances in our understanding, Es has become 'opaque', itself a somewhat opaque criticism, i.e., I'm not quite sure what it means. To be sure, few would argue with such statements as "Es as a modeling concept should always correspond to the structure, time step and spatial resolution of the model used." But to the extent that this has not been the case, it is the modeler's responsibility to define what they mean by Es, not an inherent flaw in the concept of Es itself, which is simply an attempt to define our best estimate of the average emission under a well-defined set of conditions. In the early 90's when only light and temperature were assumed to influence emissions, it was necessary to establish a specific set of light and temperature conditions under which Es was defined in order to allow comparison of emission capacity across and within species. With the discovery of additional controls over emissions - CO<sub>2</sub>, leaf age, growth environment, etc.- the definition of Es had to be expanded to include these parameters (but only when the effects of those parameters over emissions were sufficiently well characterized that a

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general algorithm could be developed to model them). To be sure, progress in incorporating such effects into existing models has been halting, not due to an 'opacity' in the concept of Es, but resulting from uncertainties concerning the generality of the effects, and inadequate mathematical representations of the effects themselves (e.g., is the effect of CO<sub>2</sub> on isoprene emissions observed in poplar a) sufficiently well-described to add to the models, and b) if so, should it be applied to all species everywhere?) In short, I believe the concept of Es is sufficiently flexible that it can, in principle, be incorporated into increasingly complex models of emission behavior, but the authors are certainly correct in suggesting that the meaning of Es varies, depending on the model being used, and that it is the responsibility of the modeler to explicitly define Es, as used in his/her application.

Virtually every published study of biogenic volatile organic compound emissions begins with a brief obligatory nod to the importance of BVOC in tropospheric chemistry, particularly their roles in O<sub>3</sub> production, determining the oxidative status of the troposphere and formation and growth of secondary organic aerosol. And many of them conclude with some variation of 'these results clearly demonstrate the importance of [Factor X] in controlling emissions of [Compound Y]; these effects must be incorporated into current emission models in order to improve their predictive capability.' Rarely do the authors provide any practical advice as to how this ought to be done. As a biologist, I'm interested in the subtleties of leaf-level emission processes, and I would never discourage basic research into better understanding of emissions at the leaf level, including biochemical and genetic controls. Sitting where I do, however, with those trying to provide a practical model of BVOC emissions to be incorporated into chemistry and transport models used by the regional and global modelers sitting one floor below, I am frustrated by the general lack of communication between the biologists providing the basis understanding of the emission processes and the end users who need a reasonable estimate of the emissions for their atmospheric chemistry models. If "Es as a modeling concept should always correspond to the structure, time step and spatial resolution of the model used," is it really reasonable to include leaf processes such as solubility

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effects, acting on the time-scale of minutes, into atmospheric models with a time step of hours? Is it necessary (or reasonable) to distinguish between monoterpene emissions from leaf pools and new production, occurring within the same leaf, in models whose spatial resolution is hundreds of square kilometers, integrating over hundreds of species? Clearly, it is important to incorporate such effects if you are a biologist attempting to understand emissions from individual leaves on short time scales, but it is naïve to suggest that large-scale emission models do the same.

It is important that the model end-users, sitting in front of their computer screens, understand at some basic level that the emissions of interest to them are the result of complex leaf-level processes, imperfectly understood. It is equally important that the biologists, sitting in front of their PTR-MS, teasing out subtle (and fascinating) biochemical explanations for variations in emissions, understand that the end-users require simply a half-hour average grid cell emission rate that captures as well as possible the integrated emissions of an infinity of leaves, to be adjusted to environmental conditions using drivers available to their models, often only light and temperature averaged over a grid cell, sometimes crude estimates of soil water.

When I speak to those modelers downstairs, they rarely ask what fraction of the modeled alpha-pinene emission is arising from storage pools. They want to know why their models overestimate observed ozone concentrations, and whether the emission models could be overestimating isoprene emissions by a factor of three. From their point of view, the subtleties don't matter; errors are much more likely to arise, for example, from inadequate representation of the amount of isoprene-emitting biomass in a given grid cell.

The authors repeatedly call for more experimental studies (to better define the Beta factor, improve our understanding of the effects of previous temperature, CO<sub>2</sub> effects, the role of non-specific storage pools & dynamic models, stresses, etc.) and I agree that such studies are required to improve our models of leaf-level emissions. Where I have a problem is with the explicit suggestion that improvement of large-scale emission

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models requires that such effects be incorporated, presumably by scaling-up leaf-level emissions to canopy-scale and ultimately to the regional scale. But if the goal is really improved emission models at the regional scale, what is needed in my view is vastly more measurements at that scale—canopy tower or aircraft flux measurements or extensive atmospheric concentration measurements above different ecosystems? Having obtained such information, observed spatial and temporal variations in emissions may be interpreted in light of information gleaned from detailed leaf-level studies.

The bias of the authors toward leaf-level investigations is evident in their list of references. There is an exhaustive bibliography of the biological aspects of BVOC production/emission obtained from leaf/branch studies, but scant reference to modeling at scales above the individual leaf, measurements of above-canopy fluxes or attempts to validate model predictions at regional scales. Of course, I'm being unfair, venting some of my own frustrations. This is clearly a review of emission controls at the leaf or sub-leaf level, and in that context, it is very useful and exhaustive. In suggesting that larger scale models are inadequate because they fail to incorporate much of the new information obtained in the last decade, however, they open themselves up to criticism.

Clearly, there is less than optimal communication between investigators at the two extremes of the continuum running from leaf biochemistry to global chemistry/transport models. In addition to being a fine review of the leaf-level end of the spectrum, perhaps this submission and the on-line dialog surrounding it can promote some discussion concerning strategies for incorporating (or not) detailed biological information into regional models, and the best ways of improving large-scale emission models in general. In any case, it is clearly useful for those end-users who rely on BVOC emission models to be reminded from time to time of the complexity and shortcomings of the models on which they rely, and the uncertainties inherent in emission model predictions. Likewise, it is important for those studying, in all their beautiful complexity, the processes leading to BVOC production/emission to put themselves in the place of both the end-user and the poor devil stuck in the middle trying to take detailed biological information and

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develop practical, large-scale, generalized models.

Some detailed comments on the text follow:

p. 1235 l. 8 delete 2nd 'in'

p. 1235 l. 22 suggest '(leaf- vs. canopy- or regional-scale, . . .'

p. 1236 l. 27 since input variables in these models are rarely on greater than an hourly time step, the model output is not an 'instantaneous' emission rate, but rather an hourly average

p. 1237 l. 4 I'm not quite sure what this means. I would define Es as the 'steady-state emission rate under a specified set of environmental driver conditions.'

p. 1237 l. 18 I think it is quite misleading to suggest, here and elsewhere, that the formulators of the original models ever considered Es to be 'a constant' for a given species. The considerable inherent variability in Es estimations was always obvious, and Es represented an average species response, based sometimes on a great many observations and sometimes on very few (or one). It is true that, once arrived at and put into the model, it is invariant, but only until someone decides upon a more appropriate value.

p. 1237 l. 25 Although it is true that different investigators have re-defined Es to incorporate additional environmental controlling factors (e.g., CO<sub>2</sub>), I'm not sure the concept of Es itself has become 'opaque'. Rather, the specific conditions under which it is estimated have changed, and it is the responsibility of each model developer to state these conditions explicitly.

p. 1239 l. 1 it is difficult to argue that "Es as a modeling concept should always correspond to the structure, time step and spatial resolution of the model used"; to the extent that this has not been the case, it is the modeler's responsibility, not an inherent flaw in the concept of Es

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p. 1243 l. 3 It is certainly true that the use of constant values for the light parameters, Alpha and CL1 ‘has no empirical justification’. The values chosen in the original publication (Guenther et al. 1993) represented the parameter values which best described the average light response measured in a few leaves each of four species. Fit individually, the values of Alpha ranged from 0.002 to 0.0036, but an average value of 0.0027 was chosen as the best practical choice to cover the range of observations. But if the authors conclude that there is no empirical justification for assigning constant values, it is incumbent upon them to suggest an alternative strategy for predicting emissions at regional to global scales.

p. 1245 l. 22 The same applies to the ‘default’ temperature response function, which represents an average response of four species grown under one set of conditions.

p. 1248 l. 17 do you mean ‘reflect’ rather than ‘affect’?

p. 1251 l. 25 “The smaller the value of H. . .” H can be in different units; what are the units being used here for H and KOW?

p. 1253 l. 8 ‘time-lags’; true, well-documented and interesting, but does incorporation of such effects really improve large-scale predictions on a hourly or less frequent time scale?

p. 1258 l. 18 ‘Es can vary widely depending on whether plants have been exposed to . . . stresses.’ There is no question that emissions vary widely depending on stress exposure; whether Es varies depends on how Es is defined. If the standard conditions under which Es is defined include the un-stressed situation, then stress effects can be incorporated via yet another dimensionless ‘stress’ scalar. As the authors point out, some limited data exists suggesting a stress dose versus emission response in some cases, which might ultimately be used to define the behavior of such a scalar. Unfortunately, as for so many of the other recommended improvements in the models, no one has put forward anything remotely practical for use in characterizing either biotic or abiotic stress levels in large-scale models.

p. 1260 l. 10 The authors are certainly correct in recognizing that, from the point of view of atmospheric chemists, it is critical to differentiate between different mono- and sesquiterpenes, given their widely varying chemical reactivities. MEGAN currently distinguishes between a number of different mono- and sesquiterpenes, but the speciation is carried out in an extremely rudimentary way, in the absence, once again, of adequate data. I certainly concur with the authors' plea that speciated terpenes be reported rather than just the sum.

p. 1266 l. 2 above-canopy fluxes will also miss any VOC deposited to the canopy while passing through

p. 1266 l. 3 above-canopy fluxes can be measured/estimated by a variety of micrometeorological techniques, from analysis of gradients to relaxed eddy accumulation (REA) to eddy covariance, and at various scales (tower, tethered balloon, aircraft).

p. 1266 l. 6 more significant that the 'tedium' involved in integrating leaf-level fluxes to arrive at a whole-canopy flux is the high degree of uncertainty involved, discussed in the previous section of the manuscript

p. 1266 l. 8 generally, eddy covariance or REA fluxes are based on integrated half-hour or hour long samples

p. 1267 l. 16 it remains to be seen whether 'the days of simple (sic) emission source modeling are past.' Anyone who has actually attempted to predict emissions from a single grid cell containing a heterogeneous mixture of emitting and non-emitting species, many of them unstudied, incorporating LAI differences, light extinction in canopies, etc., might hesitate to use the word 'simple'. Unquestionably, a great deal of valuable data relating to controls over BVOC emissions has been gathered since the early 90's and our degree of understanding has increased exponentially. Until one has tried to incorporate some of that detailed information into some sort of generalized algorithm that can be used to predict average responses of tens, hundreds or thousands of species over a large areal extent, using limited available information on environmental drivers,

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one should be careful in making such assertions.

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