

Interactive comment on “The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions” by A. B. Guenther et al.

Anonymous Referee #1

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The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions This paper marks the long-awaited update of Guenther et al. 2006, detailing for the first time the algorithms employed within the MEGAN framework for estimating emissions of compounds other than isoprene. While these algorithms are based on those presented in Guenther et al. 1995, and have been freely available to the community via the NCAR CDP data portal, this is the first time that they have been presented in print, with details

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such as emission factors and light and temperature response parameters clearly listed. This represents a substantial advance in modelling emissions of biogenic volatile organic compounds, and is timely, relevant and appropriate for publication in GMD.

The authors include an updated inventory of emissions of key biogenic trace gases from the terrestrial biosphere. The scope and intention of the paper is substantive, describing developments in both the model itself and the science behind it. As such it represents a valuable review of biogenic emissions measurements and estimates.

However, herein lies also my main concern with the paper in its present form. The authors currently devote far more time and space to a (highly informative and interesting) description of the history of research into trace gases emitted from vegetation than to the model itself. Hence, I do not feel that it meets the ethos of GMD in its current format. In addition to the concerns and comments outlined below, I would like to see the authors re-balance the paper in favour of the model itself and the algorithms employed within it. Section 3 is of insufficient length and detail as it stands. I would expect to see the algorithms employed in the model clearly presented in this paper. If it is possible to refer to other published papers for details of the majority of the algorithms in MEGAN2.1, then one might argue that this does not represent a substantial enough development or advancement of the model to justify publication in GMD.

The authors should then decide if they also intend this to stand as a review paper of emissions of VOCs from the terrestrial biosphere. If not, the first sections should be considerably reduced in scope and length. If it is intended to serve as such a review (which would also be of great benefit to the community) then not only are far more references required, but these references should reflect a greater proportion of the bVOC research community. While the group at NCAR have undoubtedly led the development of empirical algorithms for the estimation of global budgets of VOCs, many other researchers from all across the world have contributed, and have been instrumental in the flux measurements that have made this possible, and the techniques and data used to validate and constrain the output from the model.

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Other comments and concerns are outlined below.

General concerns:

(1) The authors refer repeatedly to the model's capability to estimate emissions of 147 distinct compounds. Given the considerable uncertainties in attempting to quantify even the emissions of isoprene this is disingenuous at best and misleading at worst. What is actually being presented is a model that estimates emissions of 19 compounds or compound classes, with the option to further speciate within these classes. This must be made far clearer (and far earlier) in the text, and the repeated references to 147 compounds replaced with 19 individual or lumped species or classes (precise wording is up to the authors). It should certainly not in my opinion be the "headline" claim of the Abstract. See also specific comments below.

(2) I would expect to see the algorithms/equations/response functions presented within this paper, rather than textual descriptions accompanied by references to other papers. While I acknowledge that the model itself is fully available to the community (in code format) from the NCAR data portal, to be of most use to the modelling community, this paper should act as a single stand-alone point of reference for the basic equations contained in the model. The model user should not require several other papers alongside this to piece the model together. By all means, leave some of the finer details (e.g. full details of the canopy model and the assignment of sunlit vs. shaded fraction) as further reading, but fundamentals such as the CO₂ activity factor, etc should all be set out here. Of further concern, is the authors' decision not to include details of the speciation of the 19 initial emissions groups or species into fully speciated estimates of the emissions of 147 compounds. While they have made the model's capability of estimating the emissions of 147 compounds their "headline", nowhere do they provide sufficient detail for the model user to do this.

(3) If the authors intend this to stand as a review paper as well as a model development paper, far more references are required, particularly in the earlier sections of the

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paper (1 and 2). A more diverse range (i.e. not all from the Guenther group itself) of references would also be appropriate, particularly when referring to early studies of compounds and emissions. Where possible, I have suggested starting points for references for the authors to use, but this should not be taken as an exhaustive or essential list. See also the specific comments below.

(4) While I fully agree with the authors that it is extremely difficult to quantify the uncertainties in estimates of biogenic emissions, I would expect to see an attempt to do that. This is an issue of real concern in the modelling and measurement communities and deserves a far more substantive consideration than a note in the final section of such a paper. How the authors choose to present this is up to them, (by comparison with measurements, constraints from observations (including satellite retrievals), or presenting more fully a range of estimates derived from this model) but at the very least I would expect to see a quantitative estimate of at least the order of magnitude of the uncertainty for each emission class. The uncertainties/errors inherent in the further speciation to 147 compounds should also be made clear. See also specific comments below.

Specific comments:

Abstract:

P1504, L3 - Please replace "147 biogenic compounds" with "19 compounds or compound classes, that could be further speciated to 147 biogenic compounds" or similar.

P1504, L9 - Please replace/clarify 138 compounds as above. MEGAN2.04 contained emissions algorithms for 20 compounds or compound classes.

P1504, L9-16 - The authors should make it clear that these % contributions to global annual fluxes of biogenic compounds are best estimates (or contributions calculated by MEGAN2.1). We believe as a community that biogenic emissions amount to around 1000 Pg, and that isoprene emissions are roughly half of this, etc, etc but we are uncer-

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tain of this (and even more so for the contributions of the smaller emissions classes).

P1504, L11 - please remove the word "including" as you have in fact listed all 10 compounds.

Introduction:

P1505, throughout - References are required for many of the statements in this section.

P1505, L1-2 "Terrestrial ecosystems into the atmosphere" - reference (e.g. Goldstein and Galbally, 2007)

P1505, L5-6 "...by far the biggest contributors ..." - references (e.g. Lamarque 2010)

P1505, L6-7 "...can influence atmospheric composition..." - references (e.g. Derwent et al., 2007; Folberth et al., 2006)

P1505, L9-14 "A few biogenic ... of these compounds." - references

P1505, L21-23 Please reverse this sentence as outlined above; MEGAN2.1 primarily estimates emissions of 19 compounds or classes and can then be further speciated if required, and this is how it should be presented.

P1505, L26 This sensitivity to weather and landcover data should be presented more clearly within the paper.

Section2: MEGAN compounds and sources

P1506-1522, throughout - Far more references are required for this review section of the paper.

P1506, L2-3 "...terrestrial landscapes" - reference(s) required (e.g. Goldstein and Galbally, 2007)

P1506, L3-4 "...on the atmosphere." - reference(s) required (e.g. Derwent et al., 2007)

P1506, L15-21 "MEGAN2.1 calculates light, and stress. ... (e.g. CB05, MOZART)."

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The logic of this section is inverted as outlined previously. Please put the 19 before the 147.

P1506, L21 Reference(s) required for CB05 and MOZART chemistry schemes.

P1506, L27-28 "The major ... microbes" - reference(s) required.

P1506, L28 "Humans, ... abiotic sources" - reference(s) required.

P1506, L28 "abiotic sources" - please provide some examples.

P1507, L3-5 "Foliage is thought ... for some compounds" - reference(s) required.

P1507, L13 This sentence is rather awkward, please consider revising, perhaps: "Compounds can also be released immediately after production rather than being stored."

P1507, L16 "extreme weather or herbivory." - reference(s) required. (e.g. Laothawornkitkul et al., 2009)

Section 2.1 Terpenoid compounds:

P1507, L25-26 Is there no earlier reference? 1995 is not that long ago. (e.g. Rasmussen 1978; Went 1960)

P1508, L11-13 "These activities ... plant tissues." - reference(s) required.

P1508, L22 "... a disproportionate contribution to secondary aerosol production." - reference(s) required.

P1509, L3-8 "The seventh... ..emission rates." - reference(s) required.

P1509, L11 "been observed as a component of above-canopy fluxes." - reference(s) required.

P1509, L23-25 "Capabilities (GC) techniques." Please provide references for these improved techniques.

P1510, L25 "Isoprene contributes ..." Please make it clear that this is our current un-

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derstanding based on model-generated estimates. (see e.g. Arneth et al. 2008)

P1511, L20-21 Please provide reference(s).

P1511, L22-25 Please provide reference(s).

P1512, L5 Sakulyanontvittaya et al is not a reference for high yields - please move this reference to L4 (after "SOA") and provide a suitable reference for the high yields of SOA from SQTs (e.g. Lee et al., 2006).

P1512, L14-15 "The production ... many years." - reference(s) required.

P1512, L15-17 "Two of ... Sect. 2.6." - reference(s) required.

P1512, L22-25 "Interestingly, ... beetles." - reference(s) required.

Section 2.4 Stress compounds

P1516, L20 Please add date to reference: Sawada and Totsuka (1986)

P1517, L6 "continued to emit at high rates" -> "continued to be emitted at high rates"

P1517, L10 Such observations have also been made in Europe (e.g. Ruuskanen et al 2009) - please consider expanding the references provided.

Section 2.5 Other compounds

P1518, L21-23 "These compounds ... ozone depletion." - reference(s) required.

P1519, L6-8 "Sulfur ... pristine regions." - reference(s) required.

P1520, L19-20 "These benzenoid ... to stress." - reference(s) required.

Section 2.6 Atmospheric biogenic compounds not included in MEGAN

P1521, L2-4 "Chemical species ... been identified." - reference(s) required. (e.g. Goldstein and Galbally, 2007)

P1521, L21-26 "The importance ... rates of aerosols." - reference(s) required. (e.g.

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Lelieveld et al., 2008; Vinayak, et al., 2010; Whalley et al., 2011)

Section 3 Model description:

P1524, L1 Please number this (and subsequent) equations for ease of reference

Section 3.1 Emission factors

P1524, L23-P1525, L24 "The MEGAN2.1 emission factor ... this adjustment is a few percent or less." Please could the authors clarify whether this adjustment has been made in the emission factors shown in Table 2, or whether this adjustment is specified by the model user.

P1525, L13-L18 "While it is clear... (Geron et al., 2000)" Perhaps the authors could clarify this statement. It is my understanding that observed differences in isoprene emission rates between plants of the same species grown in different locations, or even from neighbouring plants, could not be explained based on environmental factors alone (see e.g. Niinemets et al. 2010; 2011).

Section 3.2 Processes controlling emission variations

P1526-1527 (throughout) Please number all equations.

P1526-1527 (throughout) Please include the basic equations/algorithms/response functions for the activity factors shown in the equation on P1526, L7. While details of e.g. sunlit/shaded fractions/light extinction, etc within the canopy model can be left as a reference to earlier work by the authors, sufficient detail should be provided in this paper to enable model users to understand the fundamental form of the model, without reference to three other papers.

Section 3.3 Driving variables

In general, more clarity and consistency is needed in this section, in particular with regard to the consideration and reporting of the range of estimates derived from driving variables from different sources. In some cases, percentage differences in MEGAN2.1

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estimates are given, in others MEGAN2.1 estimates are compared to those derived from previous studies, and at times there is only a general statement that estimates vary according to the source of driving data or the assumptions made in its use. In part because of this lack of consistency, it is very hard to gain a clear perspective of the full range of emissions estimates from this model or to put it into context. In addition to ranges, uncertainties, etc given in the text, I would like to see this information included in tabular form; possibly two tables: one showing the range of emissions obtained from MEGAN2.1 as the driving variables are altered (much like Table 4 in Guenther et al., 2006); the second comparing the emissions of the “standard” set-up of MEGAN2.1 with estimates or ranges of emissions from previous work. This should certainly be limited to the emissions of the main compounds or compound groups.

Section 3.3.1 Solar radiation and temperature

P1527, L12-13 “... vary considerably and there is substantial range in ...” - reference(s) required. Please also quantify this considerable variation and range.

P1528, L4-5 “Substituting alternative approaches ...” Please give details of these approaches; which specific combinations of PAR conversion and radiation decomposition result in +/-30%? This could be shown in the table suggested above.

P1529, L1-5 As stated in L1 here, satellite retrievals are themselves only estimates. What uncertainty/error is associated with PAR retrieval? Please provide references to previous work comparing satellite-derived PAR and various ground-based methods of estimating PAR. Also, have the authors driven MEGAN2.1 with the satellite product referenced here? If so, what was the effect on emissions?

P1529, L6 Please change “Larger” to “Large”.

P1529, L9 Again please quantify the effect that a temperature bias such as that reported here could have on emissions estimates.

Section 3.3.2 Canopy environment

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P1529, L16 “... small underestimates in emissions.” Again, please quantify this.

P1529, L22-24 This sentence is rather awkward. Perhaps the authors could consider revising it into two separate sentences, the first stating that a better understanding is needed, and the second that this would require more observations throughout the canopy.

Section 3.3.3 Soil moisture

P1530, L2-3 References are most definitely required for this. My understanding is that some studies actually show an initial increase in isoprene emissions before decline and shutdown (followed by a further emissions spike on re-wetting). (e.g. Simpraga et al., 2011; Beckett et al., 2012; Ormeno et al., 2006)

P1530, L3 “... isoprene emission.” How do emissions of other bVOCs respond to soil moisture? The model description on P1526 gives no indication that γ_{SM} applies only to isoprene. Please make it clear whether it is intended to be set to 1 for all other species or whether the isoprene response should be considered to be representative (with references).

Section 3.3.4 PFT and LAI

P1530-1531 (throughout) Again please quantify the difference in emissions estimates resulting from the difference vegetation datasets.

Section 3.3.5 LAI

General note: could this be combined with the previous section which also considers LAI?

P1531, L21 “... substantially different ...” Please quantify the difference in emissions not just LAI.

Section 4 MEGAN2.1 emission estimates

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P1532, L18 "... will result in significant differences ..." Please quantify these differences, and include in the suggested table.

P1532, L23-4 "... we expect these estimates to have lower uncertainties." How much lower? What uncertainty would the authors estimate for the two different sets of emission factors? Please also see the general comments below for Section 5 regarding uncertainties.

P1533, L1-8 Here, the authors report % differences in emissions estimates due to different driving variables as reported in previous studies. I would expect to see a similar analysis performed with MEGAN2.1 emissions estimates: what is the range of emissions generated when changing PFT, LAI, meteorological input data, preferably in a table for clarity, in addition to the specific comments in regard to previous sections.

P1533, L8-14 "...Differences in model algorithms made a relatively small difference in global emission totals. An interesting finding ... had considerably different impacts ..." Should it not be of concern that in spite of these considerable differences, global totals appeared to be unaffected? Does that suggest robustness and confidence, or model tuning?

P1533, L25 "... similar levels of uncertainty ..." What are the levels of uncertainty in the two approaches (with references for both)?

P1534 (throughout) This section is particularly inconsistent in how differences are reported, whether they are differences within estimates from MEGAN2.1, between MEGAN2.1 and previous estimates, or ranges reported entirely in previous work (ie not associated with MEGAN2.1). Please see opening remarks to this Section.

P1534, L26 "Stavrakou et al. (2012)." Please give the estimates from this paper here.

P1535, L7-18 This is the only section of the paper that refers to and discusses these three figures. The authors should consider whether such limited use and analysis of them is enough to justify their inclusion. I would like to see them retained as I feel that

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they do add to the paper. However, in that case, far greater analysis of their implications and significance is required. For example, what processes, factors, etc give rise to the regional differences shown in Figure 4? Do we as a community have confidence in the spatial distribution of emissions of, say, β -caryophyllene shown in Figure 3? How many measurements of β -caryophyllene do we have in the tropics which are estimated to dominate global emissions?

P1535, L17-26 Surely some of the difference in total emissions is due to the considerable difference in spatial resolution of the two studies which will affect meteorological driving as well as landcover data. A lower spatial resolution of temperature or radiation implies averaging or smearing and an associated loss of extreme values which, given the non-linearity of the temperature and light response of emissions is likely to make a substantial difference in emissions estimates.

Section 5 Conclusions

P1536, L7 "... are included in a manner ..." "As previously noted, the manner of their inclusion is not explained anywhere in the paper. It should be.

P1536, L6 "Most of the 147 compounds included in MEGAN2.1 contribute very little to the total BVOC flux ..." As previously noted, a thorough and robust justification of the inclusion of so many compounds is required. While the thoroughness of the author's approach is to be applauded on the one hand (not least because it may serve to highlight the measurements or focus required in the future), it is not obvious why the authors have chosen at this stage to claim to be able to estimate emissions of 147 different bVOCs. Particularly given the lack of reliable measurements of the fluxes of most of these BVOCs, their lack of contribution to the global flux, and the lack of obvious need for such detail (how many of these are included in even the most detailed atmospheric chemistry scheme?).

P1536, L18-19 "the uncertainties associated with these emissions estimates are considerable and a lack of suitable measurements makes it difficult to even quantify these

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uncertainties.” In a paper describing the development of a widely used and respected community model, I would expect to see a thorough attempt to do that. Previous papers (eg Guenther et al., 1995, 2006, Stavrakou et al., 2011) have all tried to quantify the uncertainties associated with their models and methods, even if only to an order of magnitude. As previously outlined, I would like to see a table showing clearly the range of emissions estimates of the major compound classes included in this paper. This would go part way towards indicating the inherent variability of the estimates from this model. However further quantification of the uncertainties is clearly required if we are to be able to judge whether the development of MEGAN from v2.04 to v2.1 marks an improvement in the output of the model or merely a synthesis of the algorithms included in Guenther et al., 1995 and 2006. In addition, it must be made clear to new users of the model that the levels of uncertainty differ widely between different BVOC compound classes included in the model (presumably the authors would assume a lower uncertainty for isoprene emissions than, say, β -caryophyllene), let alone the further speciation to 147 compounds.

P1536, L21-22 “... although other compounds with a greater capacity, ...” Or that may dominate emissions in specific ecosystems or seasons?

P1537, L4 “... leaf- and canopy-scale ...” If the emission factors in MEGAN2.1 are canopy-scale emission factors, why is there a need for further leaf-level measurements to improve the model?

Tables and figures:

As noted previously, the tables and figures should be referred to and discussed far more in the text than they currently are.

Table 4 Why do isoprene, MBO and CO have values assigned for β (the light-independent exponential temperature response factor) when they have no light-independent emissions (LDF=1)?

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Table 5 The logic of the horizontal lines in this table is not clear to me. They neither split the compounds into the groups described separately in Section 2, nor the groups shown in Table 1. Should there also be horizontal lines under isoprene and MBO? Please revise to match one of Section 2 or Table 1.

Fig. 1 Please explain the significance of the different shapes and colours used for the boxes of this schematic.

Fig. 5 Please consider revising the colour scale used (perhaps the same as Fig. 3); subtle differences between shades of blue are hard to read.

A selection of suggested references:

Beckett, M., Loreto, F., Velikova, V., Brunetti, C., di Ferdinando, M., Tattini, M., Calafateira, C., Farrant, J. M. (2012) Photosynthetic limitations and volatile and non-volatile isoprenoids in the poikilochlorophyllous resurrection plant *Xerophyta humilis* during dehydration and rehydration. *Plant Cell Environment*, accepted article doi: 10.1111/j.1365-3040.2012.02536.x

Derwent, R. G., Jenkin, M. E., Passant, N. R., and Pilling, M. J. (2007). Photochemical ozone creation potentials (POCPs) for different emission sources of organic compounds under European conditions estimated with a Master Chemical Mechanism. *Atmospheric Environment*, 41(12), 2570-2579.

Folberth, G. A., Hauglustaine, D. A., Lathi  re, J., and Brocheton, F. (2006). Interactive chemistry in the Laboratoire de M  t  orologie Dynamique general circulation model: model description and impact analysis of biogenic hydrocarbons on tropospheric chemistry. *Atmospheric Chemistry and Physics*, 6, 2273-2319.

Goldstein, A. H. and Galbally, I. E. (2007). Known and unexplored organic constituents in the earth’s atmosphere. *Environmental Science and Technology*, 41(5), 1514-1521. doi: 10.1021/es072476p.

Lamarque, J. F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Li-

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ousse, C., Mieville, A., Owen, B., Schultz, M. G., Shindell, D., Smith, S. J., Stehfest, E., Van Aardenne, J., Cooper, O. R., Kainuma, M., Mahowald, N., McConnell, J. R., Naik, V., Riahi, K., and van Vuuren, D. P. (2010). Historical (1850-2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application. *Atmospheric Chemistry and Physics*, 10(15), 7017-7039.

Laothawornkitkul, J., Taylor, J. E., Paul, N. D., and Hewitt, C. N. (2009). Biogenic volatile organic compounds in the Earth system. *New Phytologist*, 183(1), 27-51. Lee, A., Goldstein, A. H., Kroll, J. H., Ng, N. L., Varutbangkul, V., Flagan, R., and Seinfeld, J. (2006). Gas-phase products and secondary organic aerosol yields from the photo-oxidation of 16 different terpenes. *Journal of Geophysical Research - Atmospheres*, 111(D17305). doi: 10.1029/2006JD007050.

Lelieveld, J., Butler, T. M., Crowley, J. N., Dillon, T. J., Fischer, H., Ganzeveld, L., Harder, H., Lawrence, M. G., Martinez, M., Taraborrelli, D., and Williams, J. (2008). Atmospheric oxidation capacity sustained by a tropical forest. *Nature*, 452(7188), 737-740.

Niinemets, U., Kuhn, U., Harley, P. C., Staudt, M., Arneth, A., Cescatti, A., Ciccioli, P., Copolovici, L., Geron, C., Guenther, A., Kesselmeier, J., Lerdau, M., Monson, R., and Penuelas, J. (2011). Estimations of isoprenoid emission capacity from enclosure studies: measurements, data processing, quality and standardized measurement protocols. *Biogeosciences*, 8(8), 2209-2246. doi: 10.5194/bg-8-2209-2011.

Niinemets, Ü.; Monson, R. K.; Arneth, A.; et al. (2010) The leaf-level emission factor of volatile isoprenoids: caveats, model algorithms, response shapes and scaling. *Biogeosciences*, 7(6), 1809-1832. doi: 10.5194/bg-7-1809-2010

Niinemets, Ü.; Arneth, A.; Kuhn, U.; et al. (2010) The emission factor of volatile isoprenoids: stress, acclimation, and developmental responses. *Biogeosciences*, 7(7), 2203-2223 doi: 10.5194/bg-7-2203-2010

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Ormeno, E.; Mevy, J. P.; Vila, B.; et al. (2006) Water deficit stress induces different monoterpene and sesquiterpene emission changes in Mediterranean species. Relationship between terpene emissions and plant water potential. *Chemosphere*, 67(2), 276-284. doi: 10.1016/j.chemosphere.2006.10.029

Rasmussen, R. A. (1978). Isoprene plant species list. Special Report of the Air Pollution Research Section. Technical report, Washington State University, Pullman, WA, USA.

Ruuskanen, T. M., Mueller, M., Schnitzhofer, R., Karl, T., Graus, M., Bamberger, I., Hortnagl, L., Brilli, F., Wohlfahrt, G., and Hansel, A. (2011). Eddy covariance VOC emission and deposition fluxes above grassland using PTR-TOF. *Atmospheric Chemistry and Physics*, 11(2), 611-625.

Simpraga, M.; Verbeeck, H.; Demarcke, M.; et al. (2011) Clear link between drought stress, photosynthesis and biogenic volatile organic compounds in *Fagus sylvatica* L. *Atmospheric Environment*, 45(30), 5254-5259. doi: 10.1016/j.atmosenv.2011.06.075

Smiatek, G. and Bogacki, M. (2005) Uncertainty assessment of potential biogenic volatile organic compound emissions from forests with the Monte Carlo method: Case study for an episode from 1 to 10 July 2000 in Poland. *Journal of Geophysical Research- Atmospheres*, 110(D23304). doi:10.1029/2004JD005685.

Vinayak, S., Williams, J., Lelieveld, J., et al. (2010) OH Reactivity Measurements within a Boreal Forest: Evidence for Unknown Reactive Emissions. *Environmental Science and Technology*, 44(17), 6614-6620. doi: 10.1021/es101780b

Went, F. W. (1960). Blue hazes in the atmosphere. *Nature*, 187(4738), 641-643.

L. K. Whalley, P. M. Edwards, K. L. Furneaux, A. Goddard, T. Ingham, M. J. Evans, D. Stone, J. R. Hopkins, C. E. Jones, A. Karunaharan, J. D. Lee, A. C. Lewis, P. S. Monks, S. J. Moller, and D. E. Heard (2011) Quantifying the magnitude of a missing hydroxyl radical source in a tropical rainforest. *Atmospheric Chemistry and Physics*,

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11, 7223-7233.

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