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Atmospheric Chemistry and Physics
Discussions

Interactive comment on “Polyols and glucose particulate species as tracers of primary biogenic organic aerosols at 28 french sites” *by* Abdoulaye Samake et al.

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The authors would like to thank Anonymous Referee #4 for his/her review and very useful comments to improve the present paper. We have studied carefully the various comments and tried to answer his/her question point by point in the following discussion.

This is a rich manuscript pooling together datasets from 28 sampling sites across France, focusing on polyols and glucose in the atmosphere. I find the manuscript to fit the scope of the journal, and to be generally well-written. I do have some fairly significant concerns about the analysis and technical comments which should be addressed

prior its publication.

General comments: 1. I find the title misleading, as there is not really use of polyols and glucose as tracers of PBOA. To achieve that, both had to be quantified and recommendations provided on how the formers can be used to estimate the latter. Instead, the manuscript is rather descriptive on polyols and glucose, and results from a largely unclear PMF analysis is given, which leads to my next comment. 2. The PMF analysis is overwhelmingly under-reported and under-explained, given that only its main results are presented. The analysis is actually referred to a report on a url which is no longer valid, or refers to publications in preparation, which is not acceptable, given that one cannot first publish the results and later the analysis. If the authors decide to keep PMF analysis for the revised version of this study, further (and complete) details of the analysis must be provided.

We agree with the reviewer that the PMF methodology was too briefly explained in the first version of the manuscript. Since PMF methodology and results are now very common in the literature, we simply referred to the European FAIRMODE guideline and to our previous papers using some of these data (Waked et al., 2014; Weber et al. 2018). We are also working on another manuscript which should described the used PMF methodology in much more details. However, based on reviewer’s comments, the revised version is now presenting in a more extended way the PMF methodology used with this large database, and associated general results. Particularly, we included a more detailed explanation of the method both in the text (please see below) and in the SI (extended text, Fig. SI-5 and Tables SI-3 to 5). Further, discussion on the statistical stability of the PBOA factor, using bootstrap analysis, is now included (Fig. SI-5) and the yearly average contribution of each PMF source-factor at each site is now provided (Fig. SI-4). The url provided for the extended report on the SOURCE program was valid, but pointed to the webpage and not to the pdf. Sorry about that. We changed this reference to point to the pdf instead: report available at https://www.lcsqa.org/system/files/rapport/lcsqa2016-

traitement_harmonise_etude_sources_pmf.pdf Regarding the title accuracy, primary sugar compounds, and in particular polyols (such as arabitol and mannitol) are now commonly recognized as suitable tracers of the PBOA emission sources (Rajput et al., 2018; Zhu et al., 2015, 2016, and therein references). In the present work, we systematically obtained a PBOA profile when concentrations of polyols (defined as the sum of daily arabitol, mannitol and sorbitol concentrations) were included in the datasets used for the PMF analysis. This PBOA profile is characterized by the presence of more than 90% of the total polyols. In this sense, we consider that we use polyols as tracers of the PBOA factor in source apportionment, and that this work is pioneering the definition of a robust chemical profile of such a factor. Further, the chemical profile, thanks to the mass fraction of these tracers, can be used to derive a mass of total PBOA. With all of these, we consider that the title is not misleading and we clarified the main text as follows: Lines 225-240, page 7: “The PMF analysis took advantage of the ME-2 algorithm to add constraints to different chemical profiles (see Tables SI-3 and SI-4 for details). Mainly soft constraints were applied in order to add some prior knowledge about the emission sources and “clean” the different profiles without forcing the model toward an explicit solution. In particular, the polyol concentrations were “pulled up maximally”, while levoglucosan and mannosan were set to zero, and EC was “pulled down maximally” in this factor in the PBOA factor. This was achieved to avoid mixing with the biomass burning factor as well as possible influences of unrealistic high contributions of EC to PBOA. Other constraints were added parsimoniously to other factors, targeting specific proxies of sources (Table SI-4). As for the general results of this large PMF study, we identified some well-known sources for almost all the sites (biomass-burning, road traffic, secondary inorganics, dust and sea salt). Two other less-common factors were identified for all sites: secondary biogenic aerosols (probably from marine origin), traced mainly by the presence of MSA, and PBOA, traced by the presence of more than 90% of the polyols total mass in the factor. Table SI-5 and Fig. SI-4 present more detailed description of the chemical tracers in each factor, together with their yearly average contribution for each site, respectively. Hereafter, only the PBOA chemical

profile will be extensively investigated. The uncertainties of this PBOA factor are discussed below and its stability is presented in Fig. SI-5. Bootstrap analysis based on 100 resampling runs evidenced the very high stability of this PBOA factor since the PBOA initial constrained factor was mapped to PBOA bootstrap factor (BF) more than 99% of the time”.

Lines 441-446, page 15: “Moreover, the sensitivity of this factor to random noise in the data was investigated thanks to randomly re-sampling the input matrix of observation. In PMF analysis, this is done via the bootstrap method (Paatero et al., 2014) in the constrained run. The PBOA factor was always mapped to itself for 13 of the sites and quasi-always (97%) for the last three ones. It means that the PBOA factor does have a very high statistical stability since it never swaps with another factor (see Fig. SI-5)”.

3. Please present your results (e.g. Fig. 6) limited only to PM10 sampling, as it is bound to represent more closely the actual atmospheric concentration, instead of being limited by too low sampling cut-off for the species studied. I recommend maintaining though section 3.3 (PM25/PM10 comparison) to report fine vs coarse mode analysis. We do agree with Anonymous Referee #4. We maintained the section 3.3, reporting and discussing the distribution of ambient polyols (and glucose) concentrations between fine and coarse mode aerosols. Following reviewer’s suggestion, we modified the Figures 6 to 8, now limited to PM10 sampling sites only.

4. I find the sampling site denominations used here unsuitable. Urban sites are typically strongly impacted by traffic emissions, so their distinction feels arbitrary. And why rural? Do you mean background? From those denominations it feels like it is lacking filter sampling at forested sites, for example. An improved description of the sampling sites is necessary to better understand its somewhat unexpected results. All of the monitoring sites (except the site of OPE-ANDRA) used in this study are stations of the air quality monitoring networks in France (AASQA). The AASQAs follow well-defined criteria for the classification (typology) of all sites in France. This methodological guide of air quality monitoring is part of the national technical reference. We kept the same

classification. Table SI-1 is also now including a web link for the description of each sampling station as provided by the respective AASQA. In this regard, it should be emphasized that urban sites presented in this study are in fact urban background sites, thus not impacted solely by traffic (in contrast to the roadside sites of Roubaix, Strasbourg, and partly Chamonix). Additionally, the rural sites are localized in rural environments, far enough from any anthropogenic sources (such as traffic, industry, etc.). This information has been added, page 5, lines 145-148.

5. There is certainly a lot to gain from combining several sampling sites, but I find that the analysis has become too shallow, unfortunately. Could you also focus on one sampling site and add more analysis (e.g. comparison with FBAP, total number, other species, wind direction/speed, backtrajectory, etc.) to try to better understand what is driving polyols and glucose atmospheric concentration? The manuscript seems to bring more questions than to answer at this point. Especially when it is kept fairly general (unclear PMF, unclear sampling periods, unclear site characterizations, etc.). We also agree with Anonymous Referee #4 that this paper is only presenting one side of a very large study, the one dealing with average concentrations of a limited number of chemical species for a large number of sites, their seasonal variations, comparison of size distributions, and chemical profile of PBOA factors obtained by source apportionment (SA) studies. This is performed by an ensemble-like study that brings, as mentioned by the reviewer, a different and robust view of the PBOA factor in the atmosphere. We found the term “shallow” a bit restrictive considering the wealth of information that is unearthed and settled by this study. We made the choice of a very different approach than that proposed by the reviewer, who is asking for more classical single site studies. It is clear that the present paper is intended to 1) settle the importance of the PBOA fraction in total PM over a large area, and 2) provide a robust chemical profile of this factor to be used in SA studies. As such, we believe that it can stand alone. However, as now mentioned at the end of the introduction, a second paper in progress will partly address the wishes of the reviewer on the processes behind the introduction of polyols into the atmosphere. And a third one will present some results

of covariation of polyols concentrations and microbial fingerprints in air, soil, and plants at a local scale. We also agree with the reviewer that the source apportionment study was lacking description in the submitted version of this paper, and it is now much more described in this updated version. We do not agree with him / her on the “unclear sampling periods” as Table S1 and Fig. S1 (already present in the submitted version) are clearly giving the dates for the sampling period at each site. We partially agree with him / her on the unclear site description. We updated the sites description table (Table S1) with web links describing each of the station, and now give a reference for the site nomenclature used for French Air Quality network. However, one should keep in mind that the characteristics of the immediate surroundings of the sites are of little importance for the atmospheric concentrations of polyols: this will be described in the second paper, showing for example that the time series of concentrations, for polyols and glucose, are within 10% of each other for 3 sites in the Grenoble area 15 km apart (one downtown in a pedestrian area, one in a park about 2.5 km away, and one in a suburban area 15 km away).

Specific/technical comments: Abstract: Unclear why dust ressuspension would be linked to PBOA factor. We do agree with Anonymous Referee #4 and have rephrased more precisely why dust resuspension could be linked to the PBOA factor. From the current literature, it is not clear if the ambient particulate polyols (tracers of PBOA) enter the atmosphere mainly through biological direct emissions or if they are associated to other materials such as the soil dust particles during resuspension processes. The contribution of crustal materials in PBOA chemical profile can give a good indication of the potential relationships between PBOA factors (polyols) and resuspension of soil dust particles. Our findings evidenced that the mean PBOA chemical profile is clearly dominated by contribution from OM (78±9 % of the mass of the PBOA PMF factor on average), with only a minor contribution from the dust class (3±4 %), suggesting that ambient polyols are most likely associated with direct biological particle emissions (e.g. active spore discharge) rather than soil dust resuspension. Our conclusions are in good agreement with those made by Jia and Fraser (2011), based on characterizing the concentrations of these chemicals in different type of samples: i.e. size-fractionated (equivalent to PM2.5 and PM10) soil, plant, fungi, PM2.5/ PM10. They found that the ambient concentrations of primary saccharide compounds at Higley (USA) are typically dominated by contributions of biological materials rather than resuspension of soil dust particles and associated microbiota.

L.53: PM affects climate, not necessarily negatively. We do agree with Anonymous Referee #4 and have rephrased this sentence (lines 53-54, page 2)

L.57: please refer to a more recent reference for carbonaceous matter. More recent references have been added (line 58, page 2).

L.57-L.66 I suggest focusing on OM on the introduction, rather than OC, an artificial species from analytical limitations. L.63: a significant fraction of OM can be associated with . . . We do agree with Anonymous Referee #4 and we focused on OM (lines 58-65, page 2).

L.72: Please specify in which environments you are referring this figure, including atmospheric layer and aerosol sizes. We rephrased this sentence and information on environment type, aerosol sizes, etc. have been added (lines 73-74, page 3).

L.74-76: And fluorescent techniques? Fluorescent techniques, in particular fluorescent microcopy methods have been previously employed to analyze airborne biological particles. This former technic has the drawback to be laborious and time consuming when it comes to analyze many samples (Bozzetti et al., 2016; Heald and Spracklen, 2009). However, it is worth mentioning that recent fluorescent techniques (e.g. UV-APS, WIBS, etc.) have considerably improve our knowledge about the abundance of airborne biological particles (Fröhlich-Nowoisky et al., 2016; Gosselin et al., 2016; Rajput et al., 2018).

L.79: Unclear how atmospheric transport complements sources and abundances The term “atmospheric transport pathways” has been removed as it can be confusing.

L.101: Datasets Large sets of data have been modified into “datasets” (line 114, page 4).

L.104: Please define atmospheric emission pathway. Do you mean the processes the plant underwent to emit polyols? More precise definition has been added. The term atmospheric “emission pathways” was used to specify how particulate polyols and glucose enter the atmosphere (line 117, page 4).

L.132: Please define “very rural”. The OPE-ANDRA site is a rural background site, which lies in the North-East of France, in a crop field area. The term “very rural” has therefore been replaced by a more precise description of OPE-ANDRA site (lines 149-150, page 5).

L.152: Please state that this number typically ranges from 1.2 to 2, so the estimates here represent an upper value of OM, thus a lower estimate of the contribution of PBOA. We do agree with Anonymous Referee #4 and this information has been added (lines 170-172, page 6).

L.185: extra space before comma. The extra space before comma has been removed.

L.186. Define JRC JRC was used to specify the European Joint Research Centre. This definition has been added in the main text (line 206, page 7).

L.194: It is unclear why mix up filter-based BC with already quantified thermo-optical EC. Or there was no EC from DECOMBIO project? Please clarify. Filter analysis for EC-OC and aethalometer measurements were conducted simultaneously for the 3 DECOMBIO sites within the Arve valley (Chevrier, 2016). Aethalometers give the total BC, thus enabling the decomposition of BC concentrations into its two main constituents: wood-burning BC (BCwb) and fossil-fuel BC (BCff) (Sandradewi et al., 2008). Considering the very specific context of this mountainous valley (with a large influence of meteorology on atmospheric concentrations in winter), the PMF runs with better results in term sof statistical stability and geochemistry were the one with BCwb and

BCff. However, for graphical simplicity, BCwb and BCff are summed up and labelled as EC in the present study. We clarified the main text as follows (lines 211-215, page 7): “The PMF conducted within the DECOMBIO project, for the sites of Marnaz, Chamonix, and Passy, used aethalometer (AE 33) measurements instead of EC (Chevrier, 2016). This complementary measure gives the total black carbon (BC), thus enabling the deconvolution of BC concentrations into its two main constituents: wood-burning BC (BCwb) and fossil-fuel BC (BCff) (Sandradewi et al., 2008). For graphical simplicity, BCwb and BCff were summed up and labeled as EC in the following figures”.

L.200: See comment #2 Please see answer at comment #2

L.211: Range values refer to min/max? In terms of readability I prefer you remove this info and present only avg±std. For readability, data has been presented as average ± standard deviation in the revised manuscript, as suggested by reviewers.

L.212: Please define Primary Sugar Compound (SC). The term Primary Sugar Compound (SC) was used to specify polyols together with primary saccharide species. The definition has been added (lines 101-102, page 3).

L.228: Please increase axis font sizes. The axis font sizes have been increased (Fig.2, page 8).

L.233: The asterisk is hard to readily identify. Please show only PM10 cutoff filters on this figure. As explained in comments #3, Fig. 6 to 8 (Fig. S5 as well) are now limited to only PM10 cutoff filters.

L.233: The selected period feels somewhat arbitrary, thus lacking a clear definition of what is shown. Please be more direct on the chosen periods (dd-mm-yyyy) and criteria applied. Selecting a specific date season could indeed be arbitrary. For simplicity, it is quite common to use months rather than the days to calculate seasonal concentration values (Verma et al., 2018). In the present work, seasons were defined as follows: Winter = December to February, Spring = March to May, Summer = June to August,

and Autumn = September to November (see lines 368-370, page 12).

Comments L.255: Please add the information of their estimated atmospheric lifetime. The primary sugar compounds (including polyols and primary saccharide compounds) are actually thought to be relatively stable in the atmosphere (Wang et al., 2018). However, studies investigating their atmospheric lifetime are quite limited. One previous laboratory study has been conducted by the US-EPA to evaluate the stability of these chemicals on filter material exposed to gaseous oxidants as well as in aqueous solutions (simulating clouds and fog droplet chemistry). Findings of this former study have shown that primary sugar compounds remain quite stable up to 7 days (the extent of the testing period), pointing out their suitability for use as tracers of atmospheric transport (Fraser, 2010). This information has added in the updated manuscript version (lines 101-109, page 3).

L.256: It feels like a weak hypothesis to me, from the PBOA perspective, could it be emission ratios change with wind speed, temperature, RH? If focusing on comparable season/meteorology, could the correlation be improved, given distinct emission pathways? And how about interferences from other sources? Is it mixing PM2.5 samples? The reviewer is right that this is a weak hypothesis, but there is very few literature on the subject (and so far no PMF study using glucose concentrations). We removed this sentence and keep working on the topic in order to figure out the sources of atmospheric glucose and its potential relation with the PBOA factor.

L.267: To improve readability, please remove SD and describe only the four average values of both sampling sites, given the interest is the distribution of fine vs coarse mode. Presenting “only” average values may certainly improve the readability. Nonetheless, average ± standard deviation give an in-depth overview of the statistical size distribution of ambient particulate polyols and glucose concentrations. In this regard, we think that it is preferable to keep these concentration values as Mean ± SD.

L.290: Please remove “compartment”. The term compartment has been removed

L.282: Please indicate the number of samples used on this analysis. The number of samples has been specified (lines 327-328, page 11).

L.301: Does it make sense that PBOA-related polyols are "only" 2-3 times higher in summer in comparison to winter time? The trend behind concentrations in "rural", "urban" or "traffic" feels inconsistent with PBOA interpretation. The seasonal concentrations of polyols presented in this section correspond to the average values over all studied sites. When focusing on one site, the summer concentrations can be 5-8 times higher than those observed in winter, which is consistent with an emission process more likely associated with increased biological activities in summertime. This observation is in good agreement with previous works (Jia et al., 2010a, 2010b; Liang et al., 2016; Verma et al., 2018). Furthermore, one should keep in mind that the values presented are seasonal averages and that daily average concentrations can be much higher in summer. Concerning the second part of the remark (about the trend according to site typology), and to the best of our knowledge, there is no observation in the literature that could support the existence of such an intuitive trend. Conversely, and as mentioned above, our measurements indicate that concentrations are nearly identical for the 3 sites of the Grenoble area, at a scale of c.a. 15 km encompassing several types of sites.

L.404: In which time series? These time series refer to a data in the study conducted by Bonvalot et al., (2016). This sentence has been removed, as the whole paragraph has been modified in the updated manuscript version.

L.440 Please correct sea-salt and not " sea minus salt". This correction has been added

L.445: Unclear sentence. This sentence has been rephrased more precisely (lines 507-524, page 17).

References Bonvalot, L., Tuna, T., Fagault, Y., Jaffrezo, J.-L., Jacob, V., Chevrier, F., and Bard, E.: Estimating contributions from biomass burning, fossil fuel combustion,

and biogenic carbon to carbonaceous aerosols in the Valley of Chamonix: a dual approach based on radiocarbon and levoglucosan, Atmospheric Chem. Phys., 16(21), 13753–13772, 2016. Bozzetti, C., Daellenbach, K. R., Hueglin, C., Fermo, P., Sciare, J., Kasper-Giebl, A., Mazar, Y., Abbaszade, G., El Kazzi, M., Gonzalez, R., Shuster-Meiseles, T., Flasch, M., Wolf, R., Křepelová, A., Canonaco, F., Schnelle-Kreis, J., Slowik, J. G., Zimmermann, R., Rudich, Y., Baltensperger, U., El Haddad, I., and Prévôt, A. S. H.: Size-Resolved Identification, Characterization, and Quantification of Primary Biological Organic Aerosol at a European Rural Site, Environ. Sci. Technol., 50(7), 3425–3434, 2016. Chevrier F (2016) Chauffage au bois et qualité de l'air en Vallée de l'Arve : définition d'un système de surveillance et impact d'une politique de rénovation du parc des appareils anciens. PhD thesis, 239 pp, https://tel.archives-ouvertes.fr/tel-01527559.

Fraser, M. P.: Final Report | Using Carbohydrates as Molecular Markers to Determine the Contribution of Agricultural Soil to Ambient Fine and Course PM | Research Project Database | Grantee Research Project|ORD|USEPA, [online]Availablefrom: https://cfpub.epa.gov/ncer_abstracts/index.cfm/fuseaction/display.highlight/abstract/7399/rep 2010. Fröhlich-Nowoisky, J., Kampf, C. J., Weber, B., Huffman, J. A., Pöhlker, C., Andreae, M. O., Lang-Yona, N., Burrows, S. M., Gunthe, S. S., Elbert, W., Su, H., Hoor, P., Thines, E., Hoffmann, T., Després, V. R., and Pöschl, U.: Bioaerosols in the Earth system: Climate, health, and ecosystem interactions, Atmospheric Res., 182, 346–376, 2016. Gosselin, M. I., Rathnayake, C. M., Crawford, I., Pöhlker, C., Fröhlich-Nowoisky, J., Schmer, B., Després, V. R., Engling, G., Gallagher, M., Stone, E., Pöschl, U., and Huffman, J. A.: Fluorescent bioaerosol particle, molecular tracer, and fungal spore concentrations during dry and rainy periods in a semi-arid forest, Atmospheric Chem. Phys., 16(23), 15165–15184, 2016. Heald, C. L. and Spracklen, D. V.: Atmospheric budget of primary biological aerosol particles from fungal spores, Geophys. Res. Lett., 36(9), doi:10.1029/2009GL037493, 2009. Jia, Y. and Fraser, M.: Characterization of Saccharides in Size-fractionated Ambient Particulate Matter and Aerosol Sources: The Contribution of Primary Biological Aerosol Particles (PBAPs) and Soil to Ambient Particulate Matter, Environ. Sci. Technol., 45(3), 930–936, doi:10.1021/es103104e, 2011. Jia, Y., Bhat, S., and Fraser, M. P.: Characterization of saccharides and other organic compounds in fine particles and the use of saccharides to track primary biologically derived carbon sources, Atmos. Environ., 44(5), 724–732, 2010a. Jia, Y., Clements, A. L., and Fraser, M. P.: Saccharide composition in atmospheric particulate matter in the southwest US and estimates of source contributions, J. Aerosol Sci., 41(1), 62–73, doi:10.1016/j.jaerosci.2009.08.005, 2010b. Liang, L., Engling, G., Du, Z., Cheng, Y., Duan, F., Liu, X., and He, K.: Seasonal variations and source estimation of saccharides in atmospheric particulate matter in Beijing, China, Chemosphere, 150, 365–377, doi:10.1016/j.chemosphere.2016.02.002, 2016. Paatero, P., Eberly, S., Brown, S. G., and Norris, G. A.: Methods for estimating uncertainty in factor analytic solutions, Atmospheric Meas. Tech., 7(3), 781–797, doi:10.5194/amt-7-781-2014, 2014. Rajput, P., Chauhan, A. S., and Gupta, T.: Bioaerosols Over the Indo-Gangetic Plain: Influence of Biomass Burning Emission and Ambient Meteorology, in Environmental Contaminants: Measurement, Modelling and Control, edited by T. Gupta, A. K. Agarwal, R. A. Agarwal, and N. K. Labhsetwar, pp. 93–121, Springer Singapore, Singapore., 2018. Sandradewi, J., Prévôt, A. S. H., Szidat, S., Perron, N., Alfarra, M. R., Lanz, V. A., Weingartner, E., and Baltensperger, U.: Using Aerosol Light Absorption Measurements for the Quantitative Determination of Wood Burning and Traffic Emission Contributions to Particulate Matter, Environ. Sci. Technol., 42(9), 3316–3323, 2008. Verma, S. K., Kawamura, K., Chen, J., and Fu, P.: Thirteen years of observations on primary sugars and sugar alcohols over remote Chichijima Island in the western North Pacific, Atmospheric Chem. Phys., 18(1), 81–101, 2018. Waked, A., Favez, O., Alleman, L. Y., Piot, C., Petit, J.-E., Delaunay, T., Verlinden, E., Golly, B., Besombes, J.-L., Jaffrezo, J.-L., and Leoz-Garziandia, E.: Source apportionment of PM10 in a north-western Europe regional urban background site (Lens, France) using positive matrix factorization and including primary biogenic emissions, Atmos. Chem. Phys., 14(7), 3325–3346, 2014. Wang, X., Shen, Z., Liu, F., Lu, D., Tao, J., Lei, Y., Zhang, Q., Zeng, Y., Xu, H., Wu, Y., Zhang, R., and Cao, J.: Saccharides in summer and winter PM2.5 over Xi'an, Northwestern China: Sources, and yearly variations of biomass burning contribution to PM2.5, Atmospheric Res., 214, 410–417, doi:10.1016/j.atmosres.2018.08.024, 2018. Zhu, C., Kawamura, K., and Kunwar, B.: Organic tracers of primary biological aerosol particles at subtropical Okinawa Island in the western North Pacific Rim: Organic biomarkers in the north pacific, J. Geophys. Res. Atmospheres, 120(11), 5504–5523, 2015. Zhu, C., Kawamura, K., Fukuda, Y., Mochida, M., and Iwamoto, Y.: Fungal spores overwhelm biogenic organic aerosols in a midlatitudinal forest, Atmospheric Chem. Phys., 16(11), 7497–7506, doi:10.5194/acp-16-7497-2016, 2016.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2018-773/acp-2018-773-AC2-supplement.pdf

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