Author Response to Referees of "Oligomer and highly oxygenated organic molecule formation from oxidation of oxygenated monoterpenes emitted by California sage plants" by Archit Mehra et al.

General Response

> We thank the reviewers for their comments for helping us to improve the paper. Referee #1 states that the interpretation of the results is sound, while offering helpful comments which have allowed us to improve the paper through suggesting ways in which we might discuss caveats to our conclusions in the light of instrumentation, calibration and experimental setup. Referee #2 states that this study is new and original, and provides interesting and fruitful results with their major concerns surrounding the experimental setup. Upon the request of Referee #2, we have added a more detailed description of the plant experiments and their measured or estimated VOC mixing ratios. With the exception of eucalyptol, the experiments were all carried out at similar VOC mixing ratios, with oligomers also observed in the plant experiments at relatively low VOC mixing ratios. We agree that the inclusion of the measured VOC mixing ratios during the single component experiments would have been a valuable addition. Unfortunately, the PTR-MS was not functioning properly during this study and we could not use any of that data, which was intended to provide us with information about the inlet mixing ratios during the single component standard experiments. We have, however estimated these concentrations, and include them in the revised manuscript. We believe that the measurements we present in this paper provide novel information on the SOA from previously uncharacterised precursors and plants and we have identified new products which may be important under atmospheric conditions. Whilst we cannot confirm their atmospheric importance, we know from the biology literature that many common plant types emit oxygenated monoterpenes. Thus, this paper serves to highlight that these systems should be considered and may be relevant for ambient SOA chemistry, and provides data for understanding their contributions in ambient measurements. Below the referee comments are in black, our responses indented and in blue, and additions to the manuscript indented and in green.

Anonymous Referee #1

Received and published: 5 May 2020

This manuscript presents measurements of VOC emissions from California sage plants and the chemical composition of subsequently formed SOA. The major finding is that highly oxygenated organic molecules (HOMs) and oligomer contribute to a larger fraction of SOA. This manuscript fits in the scope of the Atmospheric Chemistry and Physics, and the interpretation of the results is sound. While I suggest publication after major revision, I hope that the authors will consider the following comments.

1. What is the VOC concentration in the OFR? Is the high oligomer content caused by high concentrations of VOC and RO2? In other words, is the RO2 fate in the OFR representative of that in the atmosphere?

> During this study, VOC concentrations were successfully measured for the plant experiments but not for the single component experiments. Of the single component experiments, the concentration for eucalyptol can estimated to be 115 ppb by its injection rate through the syringe pump into the OFR. However, the concentrations of camphene and camphor cannot be determined by this approach as they were sublimed at room temperature into the OFR.

For the plant experiments, total VOC concentrations from the plants ranged from 27 - 34 ppb during the averaging intervals used for FIGAERO-CIMS analysis in this paper. Approximately 80 %, by mass, of the VOC emissions were oxygenated monoterpenes. Under low-NO, no seed conditions, an approximate SOA mass yield was determined of 8 % for mass loadings ranging from 4 – 19 µg m^{-3}. This value is consistent with the typical ~5-20% yields measured at similar mass loadings for other monoterpenes, such as α-pinene (Donahue et al., 2012, Figure 2a; Shilling et al., 2008, Figure 3). The SOA mass yield in this study was estimated by assuming all measured terpenes at the inlet reacted (ΔVOC, µg m^{-3}) and using AMS data for organic aerosol mass generated (ΔC_{oa}, µg m^{-3}) as SOA mass yield = ΔC_{oa} / ΔVOC. A new table has been added to the text summarizing the measured and estimated VOC mixing ratios at the OFR inlet. The aerosol mass concentrations measured during the single component studies were approximately 30 µg m^{-3} for camphor and 66 µg m^{-3} for eucalyptol. With an 8% yield, this would correspond to an estimated 60 ppb of camphor, which is still on the lower end of VOC mixing ratios used for SOA experiments historically. An 8% yield from eucalyptol would correspond to an estimated 130 ppb at the inlet, which is similar to the theoretical 115 ppb calculated based on syringe pump rate and dilution. This mixing ratio is substantially higher than the VOC mixing ratios in the camphor and plant experiments. The aerosol mass concentrations measured for camphene was 20 µg m^{-3}, to which a 12 % yield was applied based upon a literature aerosol yield of β-pinene from similar PAM-OFR experiments, owing to its structural similarity to that of camphene (Friedman and Farmer, 2018). This gave an estimated VOC concentration of 30 ppb, similar to that of the plant experiments. As highlighted in the manuscript, AMS measurements during the medium-NOx experiments had an interference and thus are not presented in the table. However, the VOC mixing ratios are expected to be the same as under low-NOx conditions, owing to the same injection conditions.

It could be the case for eucalyptol that the higher oligomer in the single component experiments was influenced by higher VOC concentrations employed in this experiment. However, camphor and camphene had similar VOC mixing ratios to that of the plant experiments alongside similar aerosol mass loadings, with camphor showing higher oligomer content which is thus unlikely to be related to VOC mixing ratios. In addition, the increased oligomer content was also observed in the plant experiments which had lower VOC mixing ratios, SOA mass loadings and SOA mass yields which suggests that the observation of oligomers was associated with the VOC itself, though contributions from the higher VOC concentrations cannot be ruled out in the case of eucalyptol.

We have added an additional section (2.5 – Summary of Experiments) containing the following text into the manuscript alongside a table of experimental conditions.

> "During this study, the VOC mixing ratios were measured for the plant mixture experiments by TD-GC-FID/ToF-MS and ranged from 27-34 ppb as shown in Table 2, with approximately 80 % by mass from oxygenated monoterpenes. Of the single component experiments, VOC mixing ratios were estimated for eucalyptol, by its injection rate through the syringe pump to be 115 ppb, while camphor and camphene could not be estimated by this approach due to their introduction through sublimation. VOC mixing ratios for camphor is thus estimated as 61 ppb on the basis of the SOA mass yields from the plants, which under low-NO, no-seed conditions which were approximately 8 % for mass loadings ranging from 4 – 19 µg m^{-3}, similar to that for other monoterpenes such as α-pinene (Donahue et al., 2012;

Shilling et al., 2008). An 8% yield from eucalyptol would correspond to an estimated 130 ppb at the inlet, which is similar to the 115 ppb calculated based on syringe pump injection rate and dilution. For camphene, the VOC mixing ratios are estimated to be 30 ppb, based on a reported yield from β-pinene from PAM-OFR experiments (Friedman and Farmer, 2018). Calculations of remaining VOC for the single component experiments shows that all VOC is reacted under under the OH exposures employed in this study, thus validating the use of aerosol yield itself to estimate VOC concentrations."

Table inserted:

Precursor	**NO_x Condition**	**VOC Mixing Ratios (ppb)**	**AMS Aerosol Mass Concentrations (µg m-3)**	**OH Exposure**		**NO ppb**	**NO_2 ppb**	**VOC/ OH**	**VOC/ NO_x**
				With VOC	**Without VOC**				
				(x 10^{11} molecules cm^{-3})					
Artemesia	Low	27*	9.35	10.02	-	0	0	48.7	
	Med	28*	-	5.29	-	2.1	51.8	97.7	0.52
Sage	Low	28*	18	7.32	-	0	0	70.6	
	Med	34*	-	4.09	-	1.7	48.5	154	0.68
Camphene	Low	30$	20	10.14	10.1	0	0	48.7	
	Med		-	4.57	4.16	2.2	58.4	121	0.49
Camphor	Low	61#	30	9.33	8.58	0	0	121	
	Med		-	4.67	3.77	2.2	57.0	241	1.03
Eucalyptol	Low	115^	66	7.16	5.51	0	0	296	
	Med		-	3.54	2.11	1.9	57.9	600	1.93

Table 1 Summary of Experimental Conditions, where VOC mixing ratios denoted by * are measured by TD-GC-FID/ToF-MS, those denoted by ^ are estimated from syringe pump injection, $ are estimated from SOA yields of β-pinene and # are those estimated from SOA yields of plant experiments .

2. The uncertainty of the contribution of HOMs and oligomers to SOA should be discussed. Firstly, the SOA product signal is represented by the sum of FIGAERO-CIMS signals. However, this by no means represents the total SOA concentration. Thus, the reported contribution likely represents an upper bound.

We agree with the referee that the sum FIGAERO-CIMS signals does not represent the total SOA concentration, and have added clarification in the manuscript that this study did not aim to relate the composition to total SOA concentration but instead compare the distribution of the SOA products detected by the different VOC systems to one another.

We have added the following text to the manuscript for clarification:

"The products detected by I-CIMS are typically multifunctional oxidised species, which do not fully represent the SOA formed from a given system. Thus the I-CIMS signal represents only a portion of the SOA and the reported contributions of this portion are likely to be an upper bound of the contribution of oligomers and HOM to total SOA. This study aims to compare the relative contributions of different SOA components across the different single precursor and plant experiments rather than quantify the absolute contributions of any of the components. Owing to the

> > similarity in composition across these systems, we expect that the ratios described here are not significantly affected by uncertainties in quantification."

Secondly, the same instrumental sensitivity is assumed for all ions. I understand the challenges in instrumental calibration, but the associated uncertainty should be considered, even qualitatively, based on previous understanding on the general relationship between compounds chemical properties and sensitivity (Lopez-Hilfiker 2016 AMT).

> We agree that the uncertainty associated with calibrations should be considered, and have added a statement discussing the potential impact of these differences:
>
> > "Iodide is more sensitive to oxidised organic species and thus is likely to overestimate the relative contribution of the more oxidised compounds and underestimate the relative contribution of the less oxidised compounds to SOA. However, given the comparison here is between VOC and oxidant systems that yield similar products, we expect that these sensitivity differences will not influence the relative contributions of different compounds classes, though estimates of absolute contributions may be an upper bound."

3. In Figure 5, how is the "% of HR signal" defined? Is this based on number or abundance? Please specify.

> This and all other figures have been changed to the same y-axis labelling of "Contribution to MS Signal (%)" for clarity.

4. Is the VOC emission profile in Figure 1 measured by I- CIMS or TD-GC-FID/ToF-MS? I would guess the latter is used, but the description in Line 175-176 ("measured with I- CIMS") is ambiguous.

> The sentence has been replaced with:
>
> > "Figure 1 shows a single snapshot of VOC composition from these plants as measured by TD-GC-FID/ToF-MS to note the main features relevant for the SOA composition measured with I-CIMS."

Anonymous Referee #2

Received and published: 25 May 2020

The authors describe a study in a PAM/OFR reactor. They oxidized emissions from two plant species and three of the major single VOC in the emissions, two of them oxidized. They studied the importance of oligomers and highly oxygenated organic molecules (HOM) for SOA formation. The focus was on oxygenated MT emitted by the Californian sage species. The study is new and original, and provided interesting and fruitful results within the scope of ACP. So the manuscript could be published in ACP. However, there are some major issues that need improvement/extension. The manuscript is relatively short so there should be room for that. In summary:

1.) Experimental details are scarce: one would need to know about the applied VOC concentrations, VOC/NOX ratios, and VOC/ oxidant ratios. I guess experiments were performed at high VOC and high oxidant concentration. In this context it should be discussed in the conclusions under which conditions and in how far the results mean anything under atmospheric conditions.

As outlined in our response to referee 1, we have added a new section titled "Summary of Experiments" which contains additional details on the applied VOC mixing ratios for the plant experiments, and includes a table of the OH exposure, VOC/NOx ratios and VOC/OH ratios the experiments. For camphor, where measurements of VOC mixing ratios are unavailable, we assume an 8 % SOA yield based upon the yields determined from the plant experiments, to estimate the VOC mixing ratios. While, for camphene, we apply a literature yield of 12 % based on that of β-pinene to estimate the VOC mixing ratios. These show that with the exception of eucalyptol experiments, the remaining experiments were carried out at relatively low mixing ratios.

We have additionally included a discussion of the remaining uncertainties with respect to the relevance of these results to ambient SOA in the conclusions:

> "This study shows that HOM and oligomer formation is potentially important for SOA formed from the oxidation of oxygenated monoterpenes. These compounds are commonly emitted by woody shrubs, but have not been the subject of previous SOA studies. Owing to the potential of these VOCs to form HOM and oligomer products, future laboratory studies should encompass a broader range of VOC/OH and VOC/NOx ratios, owing to the potential influence this has on the proportion of oligomers formed (Kourtchev et al., 2016), alongside seed acidity and humidity dependence. In addition, there are uncertainties pertaining to the absolute contributions of HOM and oligomer products to the SOA from these plant and precursor systems owing to challenges calibrating for these oxidised products for which no standards exist. These may influence the absolute magnitude of the contributions of these compound classes to SOA. This study provides insights into the SOA formation from VOC systems not previously studied for their SOA formation potential and these results should be taken as a basis upon which to study their potential importance for ambient SOA formation in more detail."

2.) **There is no information on the reproducibility or the significance of the findings, although differences between the different chemical systems are stated as results.**

This comment has been addressed in a later response where more specific questions pertaining to the significance of findings were given.

Further there is no attempt to discuss "missing calibrations" although the relative importance of different two compound classes is presented as a important result.

Though there were no direct calibrations, the relative sensitivity of the different compound classes between the different experiments would be expected to be the same. Thus, though the absolute contributions of compound classes of HOM and oligomer may be an overestimation, we feel the relative proportions of them for the different VOC systems are a valid comparison. We have added the following into the manuscript:

> "Iodide is more sensitive to oxidised organic species and thus is likely to overestimate the relative contribution of the more oxidised compounds and underestimate the relative contribution of the less oxidised compounds to SOA. However, given the comparison here is between VOC and oxidant systems that yield similar products, we expect that these sensitivity differences will not influence the relative contributions of different compounds classes, though estimates of absolute contributions may be an upper bound."

Major comments in detail: Kourtchev et al. (2016) showed that oligomer content in SOA depends on the precursor concentration and simulation experiments tend to overestimate oligomer formation compared to atmospheric situations. What were VOC/OH ratio and VOC/NOX ratio in your experiments. A table giving an overview on experimental conditions and number of replications per system will be helpful. Were experiments performed at different VOC and the results compared? If so are there any trends with VOC/OH and VOC/NOX?

As discussed above, the VOC concentrations in the plant experiments were relatively low and similar between the plant systems. In addition, the single component experiments had similar aerosol mass loadings to that of the plant experiments, with the exception of eucalyptol. Thus, though the higher oligomer content in the case of eucalyptol could be influenced by the higher VOC concentrations, the presence of oligomers in the camphor, camphene and plant experiments, where VOC mixing ratios were lower suggests that oligomer formation estimations were not significantly influenced by high VOC concentrations. We have added clarification of this to manuscript:

> "It should be noted that VOC concentrations were higher in the case of eucalyptol compared with the other single component and plant studies, and this could influence the extent to which oligomers contribute to SOA (Kourtchev,. 2016). However, the estimated VOC mixing ratios for the other single component and plant experiments were more similar to one another. Furthermore, oligomer content observed in the plant experiments which had lower VOC mixing ratios than that of the single component experiments are comparable to that of camphor."

A table has also been added to the manuscript describing experimental conditions along with a discussion of the experimental conditions, as outlined in detail in our response to referee #1, with the following text added as a result:

> "During this study, the VOC mixing ratios were measured for the plant mixture experiments by TD-GC-FID/ToF-MS and ranged from 27-34 ppb as shown in Table 2, with approximately 80 % by mass from oxygenated monoterpenes. Of the single component experiments, VOC mixing ratios were estimated for eucalyptol, by its injection rate through the syringe pump to be 115 ppb, while camphor and camphene could not be estimated by this approach due to their introduction through sublimation. VOC mixing ratios for camphor is thus estimated as 61 ppb on the basis of the SOA mass yields from the plants, which under low-NO, no-seed conditions which were approximately 8 % for mass loadings ranging from 4 – 19 µg m-3, similar to that for other monoterpenes such as α-pinene (Donahue et al., 2012; Shilling et al., 2008). An 8% yield from eucalyptol would correspond to an estimated 130 ppb at the inlet, which is similar to the 115 ppb calculated based on syringe pump injection rate and dilution. For camphene, the VOC mixing ratios are estimated to be 30 ppb, based on a reported yield from β-pinene from PAM-OFR experiments (Friedman and Farmer, 2018). Calculations of remaining VOC for the single component experiments shows that all VOC is reacted under under the OH exposures employed in this study, thus validating the use of aerosol yield itself to estimate VOC concentrations."

Experiments were not carried out at different VOC concentrations, and thus there are no trends with VOC/OH and VOC/NOx to discuss in this manuscript. As this is a potentially useful body of future work, we have included the following in the conclusions:

> "Owing to the potential of these VOCs to form HOM and oligomer products, future laboratory studies should encompass a broader range of VOC/OH and VOC/NOx ratios, owing to the potential influence this has on the proportion of oligomers formed (Kourtchev et al., 2016), alongside seed acidity and humidity dependence."

Line 206-211: Isn't that statement kind of trivial? One can only observe molecules with smaller carbon number in the particulate phase when they are more oxidized and vice versa larger molecules can make it into the particulate phase even if they are less oxidized? So what do we really learn from this? Only that camphene fragments stronger in thermodesorption?

> This discussion is relating to the relative importance of carbon backbone retaining oxidation reactions compared with those that lead to fragmentation of the carbon backbone, an additional statement has been added for clarity:
>
> > "The presence of oxidised fragmentation products in the case of camphene potentially shows that there is an increased prevalence of fragmentation pathways during the oxidation of camphene while the oxidation of camphor and eucalyptol proceed via pathways which retain their carbon backbone more readily. In eucalyptol SOA, for example, a large proportion of signal can be attributed to oxidised C10 products which form with no fragmentation of the carbon backbone, and there is a larger contribution from oligomers (≥ C11)."

Line 212: **Was the concentration of eucalyptol higher than the other species? I understand this is a laboratory study, which has its own right. Although not really stated, I guess the VOC input was high. Therefore I am wondering about OH lifetime for the (single) VOC as a measure of VOC/oxidant, and the VOC/NOX ratio. And, I am wondering in how far and under which conditions the results can be transferred to atmospheric situations?** This is also in context with the missing information on errors and significance for the discussed differences Low NOX / medium NOX, single compounds vs mixture, MT vs. oxidized MT.

> The concentrations have been discussed in previous responses, and further details of the VOC/oxidant, VOC/NOx ratio and OH lifetime are provided in the table. In addition, other comments above and discussion in the conclusions have discussed the potential relevance of these results to atmospheric situations. As the significance of the results are also highlighted in the following question, a detailed response on this aspect will be provided below.

Table 2 and line 229 -242: How many experiments were performed per VOC (mixture)? How significant are the differences, discussed in the text? Which differences are really significant? And if the differences are significant, what does it eventually mean? The paragraph is difficult to follow and should be structured clearer. E.g. I don't see some of the points made so clearly in the table.

> As is typical for chamber and OFR experiments, a single experiment was performed per VOC and plant mixture under a given set of conditions (Huang et al., 2018; Wang and Hildebrandt Ruiz, 2018; Ylisirniö et al., 2019). In this case, this corresponds to aerosol sample collected over a 20 to 30-minute period over which aerosol mass concentration and OFR conditions remain stable. The results presented in this manuscript are thus an average of the aerosol generated over this time period. Though not a true replicate, this is typical in OFR and chamber studies where it is challenging to get each experimental variable identical between experiments, for example in Table 1 in Chhabra et al., 2015. Thus the majority of studies generate SOA under various conditions just once and compare all the experiments, as we have done in this study. We have added clarification of this approach to the manuscript:

> "The intervals presented in Table 2 represents aerosol which was collected over a period of 20 – 30 minutes after aerosol mass concentrations stabilised, so each setting represents an integrated average that was analysed as a single mass spectrum. One averaging interval was used to characterize aerosol composition for each VOC system under low and medium-NOx conditions."

Regarding the significance of the differences between SOA composition, typically this is established in other fields (such as plant metabolomics) by comparing the statistical differences between the mass spectral profiles using multivariate statistical analyses (for example, PERMANOVA). However, this is not a typical approach for FIGAERO-CIMS papers published in this field with many papers comparing mass spectra in a similar manner to this study, and thus we feel that this analysis is not required for our manuscript (Huang et al., 2018; Wang and Hildebrandt Ruiz, 2018; Ylisirniö et al., 2019). In general, SOA experiments are incredibly laborious and often have low replicates, which makes multivariate statistical analyses to compare composition much more difficult than in other fields where, for example, hundreds of plant leaves can be easily sampled at one time. This is one of the primary reasons these types of analyses are not regularly performed in SOA composition studies.

We have restructured the paragraph to make the points clearer and easier to follow:

> "The table shows average O:C of I-CIMS calculated by averaging across molecular formulas and bulk O:C determined from AMS for the low NOx conditions. The average molecular formula for each condition, obtained by averaging over the distributions for each element, are also shown in the table. Table 2 indicates that increasing NOx results in a slight increase in O:C across most of the systems. The average molecular formulae indicate that this increase is caused by both a slight decrease in the average C# and an increase in the average O#."

Line 264-269: I am sorry but this comparison is empty. Didn't you made some a-pinene reference experiments under same conditions, so that you could connect your new findings with something better established?

We did not carry out experiments of α-pinene under the same conditions as the focus of this study was to characterise the SOA from novel systems. We agree that this may have helped to relate the products to something more established, however given the small contribution of α-pinene to the emission profile of the plants, we focused on those VOCs which were more abundant (camphor and eucalyptol) and unique (camphene). We understand that including this comparison with α-pinene from literature may be confusing to the reader and have thus removed it from the revised manuscript.

Section 4: Please, put your results in perspective to the atmosphere, explicitly considering the experimental conditions under which they were achieved. (A few sentences: what can be learned, where one should be careful.)

We have added a discussion in the conclusions to contextualise these results and highlight potential uncertainties:

> "This study shows that HOM and oligomer formation is potentially important for SOA formed from the oxidation of oxygenated monoterpenes. These compounds are commonly emitted by woody shrubs, but have not been the subject of previous SOA

> studies. Owing to the potential of these VOCs to form HOM and oligomer products, future laboratory studies should encompass a broader range of VOC/OH and VOC/NOx ratios, owing to the potential influence this has on the proportion of oligomers formed (Kourtchev et al., 2016), alongside seed acidity and humidity dependence. In addition, there are uncertainties pertaining to the absolute contributions of HOM and oligomer products to the SOA from these plant and precursor systems owing to challenges calibrating for these oxidised products for which no standards exist. These may influence the absolute magnitude of the contributions of these compound classes to SOA. This study provides insights into the SOA formation from VOC systems not previously studied for their SOA formation potential and these results should be taken as a basis upon which to study their potential importance for ambient SOA formation in more detail. "

Line 140, 275, and 281: I understand that calibrations are inherently difficult. However, since you are comparing two different types of SOA contribution: do you have any idea about the relative sensitivity? Please, discuss in section 2.2 your estimate and in section 4 the limitations of your findings, because of missing (possibility for) calibrations.

The following has been added to section 2.2:

> "Iodide is more sensitive to oxidised organic species and thus is likely to overestimate the relative contribution of the more oxidised compounds and underestimate the relative contribution of the less oxidised compounds to SOA. However, given the comparison here is between VOC and oxidant systems that yield similar products, we expect that these sensitivity differences will not influence the relative contributions of different compounds classes, though estimates of absolute contributions may be an upper bound."

And the following to section 4:

> "This study shows that HOM and oligomer formation is potentially important for SOA formed from the oxidation of oxygenated monoterpenes. These compounds are commonly emitted by woody shrubs, but have not been the subject of previous SOA studies. Owing to the potential of these VOCs to form HOM and oligomer products, future laboratory studies should encompass a broader range of VOC/OH and VOC/NOx ratios, owing to the potential influence this has on the proportion of oligomers formed (Kourtchev et al., 2016), alongside seed acidity and humidity dependence. In addition, there are uncertainties pertaining to the absolute contributions of HOM and oligomer products to the SOA from these plant and precursor systems owing to challenges calibrating for these oxidised products for which no standards exist. These may influence the absolute magnitude of the contributions of these compound classes to SOA. This study provides insights into the SOA formation from VOC systems not previously studied for their SOA formation potential and these results should be taken as a basis upon which to study their potential importance for ambient SOA formation in more detail."

Minor comments: In general, figures and tables need to be adapted to ACP format.

Tables and figures have been checked against ACP format, and labelling of (a) and (b) in Figure 2 changed accordingly.

Line 35: Bianchi et al. 2019 is a review, one should give the authors of the original articles also credits. I suggest to add at least e.g. Ehn et al. , Nature, 2014 here.

An additional reference to Ehn 2014 has been added to the manuscript.

Line 88: A short description of the plant-chamber and /or a reference is missing.

Inserted:

> "The plant enclosure was constructed from Teflon sheets and was approximately 45" x 14" x 14"."

Line 250 Figure 6 -> Figure 5

Changed in Line 250 and a few additional instances.

Figure 2 Venn -> Venn diagram

Changed.

Figure 2b and 3: legend should not cover information

Changed Figures.

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