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***Interactive comment on* “Better constraints on sources of carbonaceous aerosols using a combined ^{14}C – macro tracer analysis in a European rural background site” by S. Gilardoni et al.**

S. Gilardoni et al.

sgilardoni@gmail.com

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Answer to Referee #1

R: This study presents a very valuable data set, and analysis in terms of source apportionment. Such data are of great value for understanding organic aerosols in the atmosphere. The data as presented are in principal very interesting, but I think the analysis neglects some important factors. I think the authors need to re-do their QMC analysis with more realistic uncertainty bounds before this work can be published. The

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most puzzling feature of the results of this paper is illustrated very clearly in Fig.5. The summertime results show essentially zero uncertainty for EC and SOCbio. Such accuracy and precision is not possible in my opinion, especially for these components, and such results reflect the lack of appropriate uncertainty in the inputs rather than any real accuracy. A likely reason for the precision of these values is the limited attention paid to some important sources of uncertainty. Indeed, as the authors claim to be using a very efficient solver (QMC), it is hard to see why they limited their analysis to the uncertainty of just five factors (Table 2).

A: The authors are very grateful to the referee for his/her extensive comments, and specifically for pointing out the need to investigate further the effects of uncertainty sources that are not considered in the paper. In response to the main concerns of reviewer #1, we have attempted to significantly improve the uncertainty analysis, using more realistic error bounds in our Quasi Monte Carlo approach. The modifications of the text are extensively explained below.

R: Some major difficulties with interpreting this paper are:

Artifacts in the measurements themselves. For example, Gelencser et al. (2007) and Szidat et al. (2009) used error-factors for EC, which is always a significant source of uncertainty.

A: The authors introduce the measurement uncertainty of EC, OC, fM, levoglucosan, arabitol, and mannitol as an additional source of uncertainty in the QMC calculations. The following sentence is added at page 2515.

“To take into account the measurement uncertainty of EC, OC, fM, levoglucosan, arabitol, and mannitol, Gaussian curves were used to simulate distributions of observations; mean and standard deviations of the Gaussian distributions were set equal to measured concentrations and uncertainties, respectively. The uncertainties of levoglucosan, arabitol, and mannitol were calculated with the error propagation formula and taking into account peak integration, calibration, dilution error, and method reproducibil-

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ity. fM uncertainty was quantified based on analytical uncertainty. The uncertainty of OC and EC was determined based on the method reproducibility (Birch and Cary, 1996), and averaged 7% and 15%, respectively. EC uncertainty was equal to the highest estimate of the measurement artifact calculated for EUSAAR 1 protocol (Cavalli et al. 2010). ”

R: No uncertainty is allowed in the measured fM values. Although AMS instruments are renowned for their accuracy, ambient TC sampling is also renowned for its difficulty. Such filters are not always homogeneous, and the material used in the AMS does not necessarily have the same fM as the true OA at the site.

A: Uncertainty of fM is introduced as measurement uncertainty (see previous comment). In addition, replicate TC measurements on 150mm filters showed that the error associated to the filter homogeneity assumption was smaller than 4%. For 47 mm filters (as the ones used in the present study), the uncertainty would be even smaller and thus negligible compared to measurement uncertainty. The following sentence is added: “Uncertainty associated to filter inhomogeneity was neglected. In fact, replicate TC measurements on 150 mm filters showed that the error associated to the filter homogeneity assumption was smaller than 4%; for 47 mm filters (as the ones used in the present study), the uncertainty would be even smaller and thus negligible compared to measurement uncertainty.”

R: The authors restrict the range of OC to levoglucosan ratios to just 4–6 for the whole period, on the grounds that larger ratios would have led to POC_{bb} exceeding total OC. This is likely a significant source of error in summertime, as one would actually expect larger OC/levoglucosan ratios in that period. Firstly, a likely source of levoglucosan is wildfires or other open burning sources, and these have very different emission ratios to those of wintertime residential burning. Secondly, there is increasing evidence that levoglucosan can be oxidized in atmospheric conditions (e.g. Hennigan et al., 2010), and thus the measured levoglucosan may well be just a fraction of the emitted amount.

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A: The authors agree with the referee about the need of accurate knowledge of OC to levoglucosan ratio in order to model correctly the primary organic carbon from biomass burning. The ratio is expected to be higher during summer because of the different emission sources and degradation of levoglucosan. Nevertheless, the concentration of levoglucosan measured during summer was zero (within measurement accuracy) most of the time. As a consequence, using a larger range for this emission ratio would result in no changes for the biomass burning carbon concentrations. Nevertheless, according to suggestions of referee #1 and #2, the variability range of the OC to levoglucosan ratio was increased to 4-13. Text is modified as follow: "The emission ratios measured during burning of wood from central and southern Europe range between 3.7 and 12.7, while Puxbaum et al. (2007) recommend the interval 6–7 for contained combustion of European wood. In the present study the range 4–13 was employed."

R: There is no distinction between OC from BSOA and OC from PBAP, so only one value of $fM(\text{bio})$ of 1.05 is used. Although PBAP are later demonstrated to be small, the methodology would have allowed for a different value from PBAP, and the methodology would have been cleaner.

A: The authors used one single value of $fM(\text{bio})$ for biogenic primary and secondary organic carbon. Biogenic primary carbon is expected to have similar age of volatile organic carbon compounds that form secondary organic aerosol. For this reason the same reference fraction of modern carbon was employed.

R: The data chosen are a subset, chose partly to have large TC values. This obviously biases the results, and perhaps favors one type of source (e.g. anthropogenic plumes) more than others (e.g. biogenic or PBAP)? How representative were these data then? Were these the top 50% of TC samples, the top 10% or what? (I realize this is likely not possible at this stage, but it would have been useful to aggregate several low TC filters for further analysis, to get some statistics on the source of aerosol on the cleaner days.)

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A: The authors thank the referees to underline the need to better clarify the representativeness of data subset. The following sentence is added at page 2520 line 16: “The TC concentration of the subset of samples ranges from 3 to 53 ug m⁻³, while the range corresponding to the entire dataset is 0.4 – 53 ug m⁻³. The subset TC concentrations correspond to the range 15th percentile – 100th percentile, indicating that clean days were included in the source apportionment analysis, as well as polluted days.”

R: There is no tabulation of the uncertainty bounds of the different contribution estimates.

A: The uncertainties for the different contributions are discussed in paragraph 3.4 and illustrated in figure 5. Table 4 is modified to introduce source strength uncertainties.

R: The raw data are not presented, so it is impossible for others to reproduce the results given. As there are only 48 samples the measurements could easily be tabulated, perhaps as supplementary material.

A: Raw data will be made available through the EUCAARI EBAS website. In this way the scientific community will have open access to the data and, at the same time, the data authors will be informed and involved in the data usage.

R: The paper implies that the QMC method has several advantages over other methods such as LHS, but these advantages are hard to see. On p2508 it is stated that the QMC is very efficient and does not need the number of input variable combinations to be specified in advance. Does that matter? Is CPU usage for this kind of calculation worth a mention? As noted above the uncertainty analysis performed here is in fact very limited.

A: Compared to methods applied previously (such as LHS), we implemented an algorithm based on Quasi-Monte Carlo (QMC) method. To describe the advantages of this approach, text is modified as follow: “We implemented instead an algorithm based on Quasi-Monte Carlo simulations (Caflich et al. 1998); this method does not require to

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define a-priori the number of combinations of input variables, but defines this number according to the convergence of the solutions. The algorithm used gave us the option to define the probability distribution function of the input variables: for example, we used step functions to describe the variability of parameters and Gaussian functions to describe the variability of observations. Finally, the algorithm implemented in this work allowed us to vary the reference fraction of non-fossil carbon $fM(nf)$ depending on the value used for the reference fraction of biomass burning aerosol $fM(bb)$, which in turn depends on the age of the biomass burnt.

R: The terminology used for fM is confusing. For example, $fM(\text{non-fossil})$ is sometimes called the reference fraction of modern carbon in non-fossil aerosols. As is made clear by Table 2, this is not a constant, and so how is it a ‘reference’ fraction? Other papers make do with the simpler terminology fraction of modern carbon, percent modern carbon (pMC). I suggest the authors use one of the simpler terminologies, and restrict the word reference to the value used in the actual definition of fM .

A: We thank the referee for his/her comment because it points out the need to clarify the difference between reference fraction of non-fossil carbon and fraction of modern carbon. The term “reference fraction of modern carbon” is replaced by “reference fraction of non-fossil carbon” throughout the text and the following sentence is added at page 2511: “This ratio is called “fraction of modern carbon” and is denoted fM . Since it refers to the period prior to the nuclear bombing test, its value can be larger than one. Instead, the term “reference fraction of non-fossil carbon” ($fM(nf)$) indicates the factor needed to calculate the non fossil carbon concentration. $fM(nf)$ corrects for the nuclear bomb enrichment and the different content of ^{14}C of biomass burning and biogenic aerosol: because of the older age of burnt wood, carbonaceous aerosol from biomass wood burning is more enriched in ^{14}C than biogenic aerosol associated with PBAP and biogenic SOA.”

R: The comparison with model results is very thin, and wordy. Not a single figure or table is presented. Why did the authors not attempt the same type of analysis as Simp-

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son et al. (2007), who did such an analysis with the CARBOSOL source-apportionment data? They showed that one could use the discrepancy between model and observed levoglucosan to explain the total discrepancy in a convincing way, obtaining a very good time-series.

A: The comparison between model and observation monthly values is now illustrated in Figure 4. Simpson et al (2007) compared observed and modeled levoglucosan concentrations. The model underestimated the tracer concentration, as a consequence of underestimation of wood burning emissions. Similarly, the present study compares carbonaceous source strength to model TC; the underestimation of TC shows the same time trend of POC_{bb}, pointing out the underestimation of biomass burning emissions. Text is modified accordingly.

R: Abstract p2505 Line 10. The CARBOSOL project was the first long-term analysis of ¹⁴C data. The current data's improvement is to use better time resolution, so it is best to make this clear.

A: The sentence is modified as: "This was the first time that ¹⁴C measurements covered a full annual cycle with daily resolution."

R: Lines 19 onwards. Include the uncertainties in the numbers given for the different TC components.

A: Text is modified accordingly.

R: Line 21. This line (and elsewhere) refers explicitly to burning of fossil fuels, but what about OC from e.g. evaporative emissions?

A: OC from evaporative emissions would contribute to the fraction indicated as fossil secondary organic carbon. The authors thank the referee for pointing out the inaccurate use of the term "fossil fuel burning". Line 21 was rephrased as: "Moreover, aerosol associated with fossil sources represented 26% and 43% of TC" In addition, the term fossil fuel burning is replaced with fossil sources when appropriate.

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R: Line 3. There is no proof in this paper that it is just intermediate volatility products that are oxidized. Both volatile and semi-volatile VOC oxidation can lead to OA.

A: Sentence is rephrased as: “This indicates that the oxidation of semi-volatile and intermediate volatility organic compounds co-emitted with primary organics is a significant source of SOA, as suggested by recent model results

R: p2506. Line 9. Recently is a vague term.

A: “Recently,” is replaced with “During the last decade”

R: p2506. Lines 11–13. It would be interesting to see references for such a strong statement concerning OA trend in Europe. I am not aware of any good data for this.

A: Data supporting this statement are going to be presented in a publication in preparation. The sentence is rephrased as:” Long-term measurements have shown that in Ispra the European legislation has effectively succeeded in reducing PM10 concentrations over the last decade (Gruening et al. 2009), while the relative contribution of carbonaceous aerosol to PM2.5 increased (Yttri et al. 2009). “

R: p2506. Lines 25–27. Make it clear that the results of Zhang et al. are global. What about European data - there are plenty of AMS results available now.

A: Sentence is rephrased as:” Field measurements on global (Zhang et al. 2007) and European scale (Morgan et al. 2010, Lanz et al. 2010) have shown that oxygenated organic aerosol (OOA), of which a major fraction is SOA, composes on average 60% of submicron organic mass in urban locations and 80% downwind of urban areas.”

R: p2507. Lines 1–3. This sentence is unclear. There are many tracer methods, and many of these do not use chemical profiles (for example, 14C is a common tracer) Kleindienst et al., (2007) used tracers but not really profiles - the latter word is usually associated with CMB type analysis.

A: Sentence is rephrased as: “Receptor models, like Chemical Mass Balance, ap-

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portion primary carbonaceous aerosol sources based on the knowledge of chemical profile of each single source and the unapportioned mass is then assigned to SOA”

R: Line 9. Why say “seldom verified”? This methodology is fraught with weaknesses, and so “verification” would be impossible.

A: Sentence is replaced by: “This last assumption is seldom justified,”

R: Lines 14–15. Better to reference here the more pioneering studies of Szidat et al., 2004, Gelencser et al., 2007. Last line. The $^{14}\text{C}/^{12}\text{C}$ ratio also depends on the age of the modern carbon sources. This is explained later, but should be mentioned when this ratio is first introduced.

A: Pioneering work describing the use of ^{14}C to quantify biogenic aerosol is represented by Currie et al (1982). References are added and the sentence is modified as “ the $^{14}\text{C}/^{12}\text{C}$ ratio in fossil fuels is zero and the isotopic ratio of atmospheric aerosol depends on the relative contribution of fossil and non-fossil carbon and on the age of modern carbon sources.”

R: p2508. QMC is introduced here, but no references are given, which makes it hard for the reader to understand what is being used (I doubt many are familiar with the term QMC).

A: The following reference is added: Caflich, R. E. Monte Carlo and quasi-Monte Carlo methods, Acta Numerica, 1998, 1-49.

R: p2508, lines 20 on - it is stated that the paper introduces uncertainty of two values, the reference fraction of aerosol modern carbon and biomass burning aerosol. This sentence makes no sense at this stage of the manuscript, and is not very clear even after reading the rest.

A: The sentence is rephrased as: “Finally, the algorithm used in this work allowed us to vary the reference fraction of non-fossil carbon ($f_{\text{M}}(\text{nf})$) depending on the value used for the reference fraction of biomass burning aerosol ($f_{\text{M}}(\text{bb})$), which in turn depends

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on the age of the biomass burnt.”

R: p2510, line 17 ‘the atmospheric stability of levoglucosan was verified’. This is a strong statement, especially given the mounting evidence that levoglucosan can degrade in the atmosphere. A reasonable correlation with K (and just $r^2=0.73$) might imply no more than that the sources are close by. Indeed, a correlation of 0.73 between compounds can be easily found between reactive and non-reactive gases, if subject to similar meteorology and source-areas.

A: Sentence is rephrased as: “the correlation of levoglucosan with non-dust soluble potassium, an inorganic tracer of biomass burning ($r^2 =0.73$), confirms that atmospheric degradation, if present, did not compromise the use of this marker as a specific tracer of biomass combustion.”

R: p2511, line 17. CO₂ did not double ‘momentarily’.

A: Sentence is corrected as: “CO₂ almost doubled during the tests.”

R: p2511, last two lines. As the concept of standards for modern carbon and fM are often confusing, it would be good to make clear here that fM values can exceed one, and explain why.

A: Sentence is modified as:” This ratio is called “fraction of modern carbon” and is denoted fM. Since it refers to the period prior to the nuclear bombing test, its value can be larger than one.

R: p2512. The methodology used here follows quite closely that suggested by several other workers, e.g. Szidat et al or Gelencser et al. It would be fair to mention this at the beginning of section 2.4.

A: The following sentence is added in the introduction: “This study presents a similar approach to that suggested by Gelencser et al. 2007 and Szidat et al. 2009, who used Latin Hypercube Sampling to calculate a large number of combinations of the input variables and to solve the problem.” Gelencser et al. 2007 and Szidat et al. 2009 are

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quoted again at the beginning of section 2.4.

R: p2512, line 21. Why not consider modern carbon emissions in summertime from other sources, e.g. grilling? What about other biofuels?

A: As stated at page 2512, the study assumes that biomass burning aerosol, fossil sources and biogenic sources describes the major sources of carbonaceous aerosol in the area.

R: p2513, line 16. When stating that something is negligible, give a reference.

A: The reference is given at the end of the following sentence. For clarity the punctuation is modified as follow: "Pollen grains, fragments of plants and animals are typically larger than 10 μm and their contribution to fine aerosol is negligible; on the contrary, spores can be smaller than 10 μm , bacteria can be as small as 1 μm , and viruses range around 100nm (Matthias-Maser and Jaenicke, 2000)."

R: p2513, lines 18–19. The authors claim that only fungal spores can contribute significantly to non-forested areas. First, what do the authors mean by significant? Puxbaum and Tenze-Kunit found significant contributions from plant-debris in downtown Vienna, and Gelencser et al found between 1–8% contributions across the CARBOSOL sites.

A: Sentence is modified as follow: The concentration of primary biogenic has been quantified using cellulose as a tracer of vegetation debris (Sanchez-Ochoa et al. 2007) and saccharides as tracers of fungal spores (Bauer et la. 2008a,b, Jia et al. 2010). In the present study we quantified primary biogenic OC based on arabitol and mannitol concentration, assuming that fungal spores dominated PBAP. Nevertheless, it should be kept in mind that other PBAP could be present and this work calculated a lower estimate of primary biogenic OC."

R: p2513. Why choose the lower bound of 5.2 pg C/spore from Bauer et al.? As the QMC method can look at a range of possibilities, why not specify the range and let QMC work with that.

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A: The content of OC from a single spore was assumed equal to 5.2 pg C spore⁻¹, which corresponds to the lower bound reported for PM₁₀ aerosol (Bauer et al. 2002), because this study focus on PM_{2.5} fraction and the use of a larger OC content would be unrealistic. Sentence is modified as follow: “To calculate the concentration of POC_{bio} in fine particles (PM_{2.5}) the content of OC from a single spore was assumed constant and equal to 5.2 pg C spore⁻¹ corresponding to the lower bound reported for PM₁₀ aerosol (Bauer et al. 2002).

R: p2513. It isn't clear if both mannitol and arabitol are used, and how

A: The following sentence at page 2513 is corrected:” The number of spores was calculated as the average number of spores obtained from arabitol and mannitol concentrations, according to Bauer et al. (2008a).

R: p2514. Why does biomass burning get reduced to bb in fM(bb), whereas non-fossil is fully spelled out. Be consistent (and I suggest using the shorter versions in subscripts).

A: The term fM(non-fossil) is replaced with fM(nf) throughout the text.

R: Why do IVOCs get a mention, but not SVOCs? Doesn't POC_{ff} as derived from ambient studies also include some oxidized IVOC?

A: Sentence is corrected: “SOC_{ff} and SOC_{bb} correspond to OC produced by the oxidation (e.g. through aging) of intermediate volatility (IVOC), semivolatile (SVOC), and volatile organic compounds (VOC) from anthropogenic activities (Robinson et al., 2007),” The presence of IVOC in POC_{ff}, and consequent overestimation of POC, is due to the use of EC to OC ratios that are not representative of ambient conditions, and it is discussed at page 2518 line 1.

R: fM(bio) is only known if one assumes zero PBAP.

A: There is no reason why PBAP should have reference fM different from that of biogenic secondary organic aerosol.

R: p2515. I think the authors are basically applying a top-hat distribution to their inputs, between the min and max values. Is that correct?

A: The distribution of inputs is clarified at page 2515 line 11. “and the 5 input parameters were allowed to vary with a step function across the whole uncertainty range reported in Table 2.”

R: p2517, line 2 - what is the question mark before Schmidl?

A: Text is corrected.

R: p2520. The ratio of OM:OC is too low for the type of ambient sample discussed here. Indeed, later in the manuscript the authors themselves use 1.8, although for some reason using data from Amazonia. Turpin+Lim (2001) is still probably the classic paper on this issue.

A: At page 2520 we calculated the OM concentration for the entire dataset using the OM to OC ratio of 1.4, obtaining a lower estimate of the OM concentration. The purpose of this calculation is to show the importance of organic mass relative to the fine aerosol mass. The average OM of the subset of samples is calculated using again the ratio 1.4 for consistency. A more accurate calculation of OM is presented in section 3.3 and compared with the lower estimate. The following sentences are added.

“Organic mass (OM) was calculated assuming an OM to OC ratio equal to 1.4; this value, which is close to the lower bound reported in literature (Turpin and Lim 2001, Aiken et al. 2008), leads to a lower estimate of the OM contribution to fine aerosol mass. Nevertheless, OM represented 53% and 41% of fine mass during winter and summer, respectively, while the EC contribution was 10% and 7%.”

“AMS studies reported OM to OC ratios in the range 1.4 - 2.5, with higher values for oxidized organic aerosol and smaller values close to the emission sources (Aiken et al. 2008). In this study the ratio 1.4 was used for POA, and 1.8 for SOA.”

“The average OM, calculated as sum of SOA and POA, was 30.2 $\mu\text{g m}^{-3}$ and 10.6 $\mu\text{g m}^{-3}$ ”

m-3 in winter and summer, respectively. These values are 10 to 17% higher than the OM estimates presented in section 3.1, indicating that an accurate choice of the OM to OC ratio has to take into account the aerosol sources and their seasonality.”

R: p2523. Although the discussion of Fig. 4 is very interesting, it is a stretch to explain the good correlation between SOC_{bio} and POC_{ff} only in terms of condensation effects. Meteorology is very good at generating good correlations between parameters. This could be checked by comparing correlation with other (non-carbonaceous) pollutants.

A: Figure 4 is removed. We agree with the referee that dilution may lead to correlation of atmospheric pollutants. SOC_{bio} and POC concentrations were normalized to fine mass in order to remove the effect of atmospheric dilution. Text is modified as follows: “Daily samples collected during the same months showed that the mass fraction of biogenic secondary carbon (i.e. SOC_{bio} normalized to the fine aerosol mass) correlated very well with the mass fraction of primary carbon (i.e. the sum of primary OC and EC normalized to fine mass), that in summer is emitted almost exclusively by anthropogenic fossil sources. The normalization to fine mass is used to remove the effect of meteorology. SOC_{bio} has different sources (oxidation/aging of biogenic volatile precursors) compared to primary organic particles (combustion processes, transportation). PSCF (section 3.6) indicates that the source region of the two carbon fractions are different: primary fossil carbon is produced by local sources and emissions of urban and industrial areas in the nearby Po valley, while biogenic SOA is produced by oxidation of biogenic volatile compounds, likely originated in the rural area around the sampling site. In addition, the lack of correlation among other carbonaceous particle fractions indicates that residence time or similar history of carbonaceous particles is not enough to explain the relationship between biogenic secondary and primary OA. In conclusion, the good correlation between SOC_{bio} and primary carbon fractions ($r^2 = 0.79$) could be explained by the promotion of biogenic secondary aerosol formation by primary carbonaceous material. As suggested by Bowman et al. (2004), a larger fraction of anthropogenic aerosol can offer a larger surface area with chemical affinity

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for condensation of biogenic gas phase precursors.”

R: Table 2 - add the k1..k4 nomenclature here

A: Table is corrected

R: Table 3. It isn't clear what is meant by non-null values. If concentrations are measured to be zero (or below the detection limit) then this is a valid observation and methods exist to include these in the averages. As it is, the values given for e.g. OC cannot now be compared with those for levoglucosan or mannitol. Of course, if the authors list all their data as suggested above this difficulty is partly removed.

A: The average values are recalculated using the substitution method suggested by Antweiler et al. (2008). Values below detection limit are replaced by one half of the detection limit value.

R: Table 4 should include the uncertainty estimates.

A: The uncertainties are discussed in section 3.4 and reported in figure 5. Table 4 is modified to introduce source strength uncertainties.

R: Fig. 1: the lines are hard to see. Maybe thinner lines with different styles would help.

A: Figure 1 is redrawn.

R: Fig. 4 could be merged with Fig. 6 or even omitted.

A: Figure 4 is omitted.

References

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