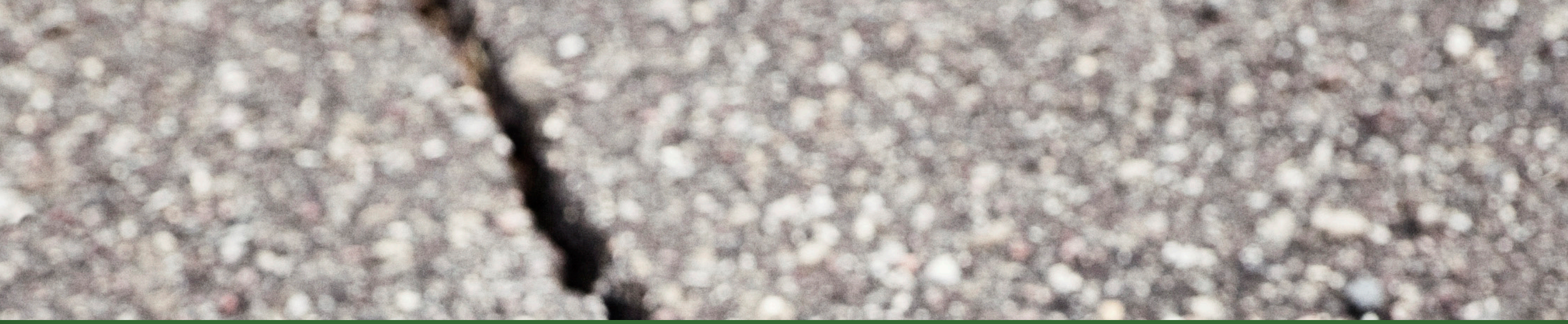

Re-Road *End of life strategies of asphalt pavements*

Sampling and Characterization of Reclaimed Asphalts – Final Report

Virginie Mouillet et al.

SEVENTH FRAMEWORK PROGRAMME

The research leading to these results has received funding from the European Community's Seventh Framework Programme (FP7/2007–2013) under grant agreement n° 218747.

Re-Road – End of life strategies of asphalt pavements

Deliverable 1.7
Final report
“Sampling and characterization of Reclaimed Asphalts”

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EUROPEAN COMMISSION
DG RESEARCH

A FP7 Collaborative Project
Work programme:
Sustainable Surface Transport
SST.2007.1.2.2 End of life strategies
for vehicles/vessels and infrastructures

Executive summary

This deliverable synthesizes the findings which are discussed in detail in deliverables D1.2 "Methodology for laboratory characterization of Reclaimed Asphalts" (Mouillet et al., 2011), D1.3 "Sampling procedure for Reclaimed Asphalts" (Gabet & Wayman, 2012), D1.4 "Test procedures/methodologies for Reclaimed Asphalts binder" (Mouillet et al., 2012), D1.5 "Test procedures/methodologies for quickly screening of Reclaimed Asphalts" (Gobert et al., 2012) and D1.6 "Guide for environmental characterization of Reclaimed Asphalts" (Enell et al., 2012). Further some significant results related to this work Package 1 but obtained in the framework of work packages (WP2 and WP3) are also discussed in this report and drawn for conclusions. As an executive summary, the significant results are presented in section 2.

This report discusses following topics in detail:

- How to best sample RA.
- Improvement of the technical characterization of RA, especially RA containing Polymer modified Bitumens.
- Definition of an appropriate methodology to better know the pollutant potential of RA.

Finally, recommendations are given for the optimal sampling of RA in mixing plants for further characterization using laboratory methods to assess the quality of RA in physical, chemical and environmental domains. Relevant indicators for estimating the recycling potential of a RA are also proposed.

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List of abbreviations

AC	Asphalt Cement
ARA	Asphalt containing Reclaimed Asphalts
BC2	Bucklet centrifuge type 2
BSM	Benzene Soluble Matter
CFC	Continuous Flow Centrifuge
DCM	Dichloromethane
DOC	Dissolved Organic Carbon
EVA	Ethylene-Vinyl Acetate
FTIR	Fourier Transform Infra-Red spectroscopy
HOC	Hydrophobic Organic Compounds
HPCD	hydroxypropyl-β-cyclodextrin
PA	Porous Asphalt
PAH-16	naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene dibenzo(ah)anthracene benzo(ghi)perylene and indeno(123cd)pyrene
PAH-L	Polycyclic Aromatic Hydrocarbons with low molecular weight (naphthalene, acenaphthylene and acenaphthene).
PAH-M	Polycyclic Aromatic Hydrocarbons with medium molecular weight (fluorene, phenanthrene, anthracene, fluoranthene, pyrene).
PAH-H	Polycyclic Aromatic Hydrocarbons with high molecular weight (benzo(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene dibenzo(ah)anthracene benzo(ghi)perylene and indeno(123cd)pyrene).
PCE	Perchloroethylene
PmB	Polymer modified Bitumen
RA	Reclaimed Asphalts
R&B	Softening point
SBS	Styrene-Butadiene-Styrene
SMA	Stone Mastic Asphalt
SVOC	Semi Volatile Organic Compounds

TCE	Thrichloroethylene
TOCe	Total Organic Carbon emitted
Tol	Toluene
$T_{R\&B}$	Ring&Ball Temperature

1 Introduction

1.1 Overview Re-Road WP1

The vast majority (90%) of European roads are paved with asphalt material. At the end of the service lifetime of a road, when the damaged pavement cannot further fulfil its purpose as a comfortable carrier of traffic, the road pavement must be renewed. Sustainable construction processes that conserve natural resources are well recognized within the asphalt industry, although practices for asphalt recycling vary to a great extent across Europe. Today a large amount of demolished asphalt pavement ends up as unbound granular layers where neither the bituminous binders nor special aggregates from old surface layers are reused at their full potential.

The Re-Road project aims to develop knowledge and innovative technologies for enhanced end of life strategies for asphalt road infrastructure. Such strategies have an important positive impact on the energy efficiency and the environmental footprint of the European transport system and fit within the life-cycle thinking which is being introduced in waste policy at European level.

One of the topical objectives of the Re-Road project is to allow re-use of the optimal proportion of recycled asphalt pavement in wearing courses, according to industrial means and costs for recycling. In this framework, the aim is to re-use higher amounts of Reclaimed Asphalts (RA) in mixes in comparison to current practices.

However, RA are complex materials and the use of significant proportions of RA involves a more critical management of heterogeneity and a necessarily more accurate control of the characteristics of these materials. Consequently, the WP1 focused on issues that are specifically related to the characterization and technical evaluation of RA as a raw material prior to the recycling processes. The main goal of this WP1 was to better assess the characteristics of RA that will allow an increase of the percentage of RA in new asphalt mixtures while ensuring final material performances. Further, in the framework of sustainable development, the use of higher amounts or RA will permit to reduce natural aggregates consumption and to decrease storage sites for wastes.

The WP1 research program involved 9 European partners; it covered the following topics that are important for improvement of RA characterization in laboratory and for assessment of the potential of RA (binder & aggregates) to be recycled:

- Development of a suitable sampling procedure to consider heterogeneity of materials that could be an important source of uncertainty for the assessment of physical, chemical and environmental properties of RA or materials including RA. One of the most important factors influencing the heterogeneity is the sampling. So, the aim of the task 1.1 "Sampling procedure for RA" was to deal with the improvement and/or development of an optimal sampling strategy for RA in mixing plants, taking into account further characterization.

- Proposition of methods of RA assessment in physical and chemical domains. The composition of RA is an important issue related to the possible re-use for these materials. In order to improve mechanical performances, bituminous materials, especially for wearing courses and for thin surface layers, include more and more Polymer modified Bitumens (PmB). When these materials are recycled, their characterization is difficult because of the lack of suitable test methods for their analysis and/or for the determination of their ageing level. Also, their impact on performance is unknown. Moreover, in order to re-use RA in surface layer or to choose a more appropriate road layer, it is necessary to identify different aggregates parameters, like shape and intrinsic characteristics. Consequently, the goal of the task 1.2 "Improvement of the characterization of RA, especially RA containing Polymer modified Bitumen (PmB)" dealt with the characterization of the bitumen part and the granular part of the RA, in order to take into account actual means and their (dis)advantages for studying RA.

- Evaluation of test methods for environmental characterisation of RA. For RA to be successfully reused in high value recycling applications, such as reuse in new asphalt surfaces, it is necessary to ensure that the environmental characteristics of the material are fully understood. Key tools to help gain this understanding are risk assessment and life cycle analysis of the various stages of the recycling process (Enell et al., 2012), however, to allow for meaningful analyses it is important that these analyses are based on relevant and valid test data. An important task of WP1 was hence to search for appropriate methods that could quantify the emissions of potential hazardous compounds arising from RA. This work was organized under task 1.3 "Environmental characterization", which focused on identification and evaluation of methods intended for quantification of water- and airborne emissions. In additions characterisation tests that can support the environmental assessment of RA, such as ecotoxicological and bioavailability tests, was also included for evaluation.

1.2 Partners/Authors

This deliverable summarises the research work conducted for the Re-Road project, work package (WP) 1. Following authors and partners contributed to the experimental studies and discussions of the presented results:

- Virginie MOUILLET (Ifsttar), WP1 leader and Task Group 1.2 leader
- Thomas Gabet (Ifsttar), Task Group 1.1 leader
- Anja Enell (SGI), Task Group 1.3 leader
- Konrad Mollenhauer (TUBS)

- Nathalie Piérard (BRRC)
- Ola Wik (SGI)
- Vincent Gaudefroy (Ifsttar)

Additionally, the technical staff conducting the experiments at the participating laboratories is acknowledged.

1.3 Scope of the report

This deliverable synthesizes the findings which are discussed more in detail in the six deliverables produced in the framework of Work Package 1 :

- ❖ D1.1 "State of the art on existing laboratory methods (sampling, characterization) linked to Reclaimed Asphalts study"" (Mouillet et al., 2010),
- ❖ D1.2 "Methodology for laboratory characterization of Reclaimed Asphalts" (Mouillet et al., 2011),
- ❖ D1.3 "Sampling procedure for Reclaimed Asphalts" (Gabet & Wayman, 2012),
- ❖ D1.4 "Test procedures/methodologies for Reclaimed Asphalts binder" (Mouillet et al., 2012),
- ❖ D1.5 "Test procedures/methodologies for quickly screening of Reclaimed Asphalts" (Gobert et al., 2012),
- ❖ D1.6 "Guide for environmental characterization of Reclaimed Asphalts" (Enell et al., 2012).

Further some significant results related to this work Package 1 but obtained in the framework of work packages (WP2 and WP3) are also discussed in this report and drawn for conclusions. As an executive summary, the significant results are presented in section Executive summary of WP1 results.

This report discusses following topics in detail:

- How to best sample RA.
- Improvement of the technical characterization of RA, especially RA containing Polymer modified Bitumens.
- Definition of an appropriate methodology to better know the pollutant potential of RA.

Finally, recommendations are given for the optimal sampling of RA in mixing plants for further characterization using laboratory methods to assess the quality of RA in physical, chemical and environmental domains. Relevant indicators for estimating the recycling potential of a RA are also proposed.

2 Executive summary of WP1 results

Reclaimed asphalts (RA) are complex materials. Their characteristics are difficult to assess. Thus, their use in new asphalt layers in significant proportions presents certain complications. It requires better management of heterogeneity and close management of RA characteristics. Consequently, WP1 focused on issues that are specifically related to the characterization and technical evaluation of RA as a raw material prior to recycling processes. The main purpose of WP1 was to better assess the characteristics of RA to promote an increase in their proportion in future asphalt mixtures whilst maintaining final material performances. Therefore, specific topics were elaborated in three task groups:

- ✓ Task Group 1.1: Sampling procedure for RA,
- ✓ Task Group 1.2: Improvement of the characterization of RA, especially RA containing Polymer modified Bitumens (PmB),
- ✓ Task Group 1.3: Environmental characterization.

The results of each task groups are presented in deliverables D1.2, D1.3, D1.4, D1.5 and D1.6 on which this synthesis report is based on. The following section summarises the results obtained in the single work tasks. For details, the deliverables are accessible online.

2.1 Results obtained in Task Group 1.1

The first task of WP1 was dedicated to the development of a suitable sampling procedure, which considers heterogeneity of materials that could be an important source of uncertainty in the assessment of physical, chemical and environmental properties of RA or materials including RA.

In the first deliverable dealing with the state of the art (D1.1), a synthesis of knowledge and practices of WP1 partners on issues related to the characterization and technical evaluation of RA prior to the recycling processes was performed. In task group 1.1, a sampling procedure has been proposed, based on existing European standards and practices of different countries. It was agreed that the aim of the sampling procedure was to assess the highest level of recycling for a RA, but the procedure should be adapted to the kind of material and the way it is stored. Indeed, in the case of unknown materials stored and mixed in a unique stockpile, it can be expected that the material will be re-used only at low level of recycling. However, it seems impossible to characterize such kind of stockpile in terms of heterogeneity. In that case tests of characterization would be too fastidious to perform, for time, money and technical considerations. On the contrary, when RA is well-known, when it comes from a unique source, and when it is stored in a homogenous stockpile, it is expected that the material can be re-used at a high level of recycling. Then, a higher number of samples will be needed to characterize this RA.

Consequently, the proposed procedure is the result of a compromise solution that considers an expected level of recycling as a function of the origin of RA, the kind of stockpile and the initial data on the material. According to these different parameters, a level of recycling and its associated level of characterization can then be proposed. Sampling and characterization will be used to confirm the expected level or impose a lower level of recycling if results of tests do not reach the required quality. Now that the main principle behind the procedure has been established two issues remain to be solved:

- The first issue is to set practical values to define the level of recycling as a function of the cases. This issue still needs studies.
- The second issue concerns restricting the level of recycling for unknown mixed RAs, despite the possibility that they could be of a good quality, according to some stockpile owners. Due to a lack of time and study cases, these questions did not fall within the scope of this task group 1.1.

However, part of this procedure was investigated within the scope of the deliverable D1.3. In the sampling procedure proposed in deliverable D1.1, three cases were considered (see Figure 1).

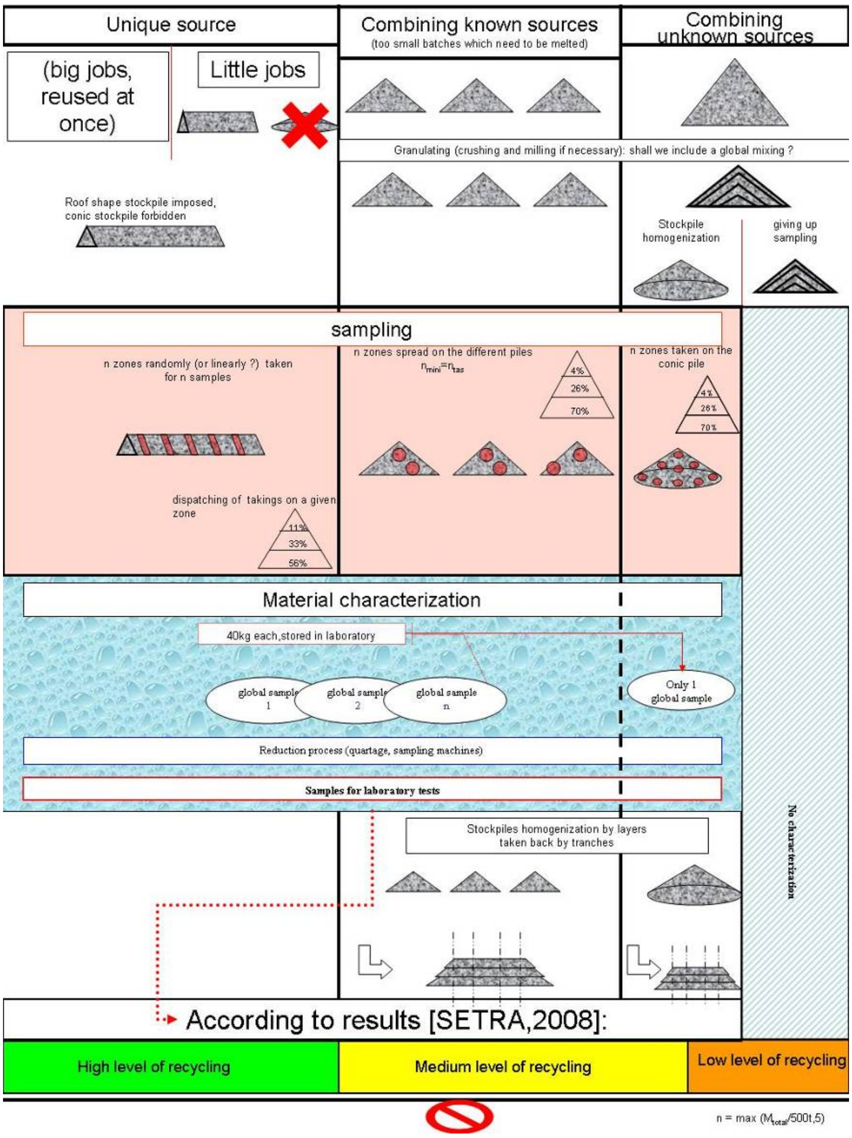

Figure 1: A sampling procedure adapted to kinds of RA and stockpiles and levels of recycling.

The first case (described on the left of the Figure 1) concerns well known stockpiles arising, for example, on big job sites like motorways where large sections of old courses are produced as RA. In that case, sampling and then characterizing RA seems feasible in the sense that material is often well-stored and homogenous. Sampling based on the present standards (EN 13108-8 and EN 932-1) is sufficient to characterize this type of material. In the third case, which requires only a little (or none) characterization of RA, is also not complicated to deal with. The work of task group 1.1 focused on the second case, consisting of mixing known sources (considering that the owner has made the effort to store separately its RA!), and their possible use at a medium level of recycling. Mixing RA sources in this manner requires characterization of the mixture. In that case, it has been thought that mixing laws could been used to estimate the characteristics of the mix, while avoiding to perform further tests. Mixing laws are usually used to estimate the properties of a mix of binders. Mixing laws used in that study were as following:

- For the binder content of the mix, a linear law was used:

$$bc_{mix} = \frac{M_1.bc_1 + M_2.bc_2 + M_3.bc_3}{(M_1 + M_2 + M_3)}$$

- For the penetration value of the mix, a logarithmic law was used:

$$\log(pen_{mix}) = \frac{M_1.bc_1.\log(pen_1) + M_2.bc_2.\log(pen_2) + M_3.bc_3.\log(pen_3)}{(M_1.bc_1 + M_2.bc_2 + M_3.bc_3)}$$

- For the Ring and Ball temperature, a linear law was used:

$$T_{mix}^{r\&b} = \frac{M_1.bc_1.T_1^{r\&b} + M_2.bc_2.T_2^{r\&b} + M_3.bc_3.T_3^{r\&b}}{(M_1.bc_1 + M_2.bc_2 + M_3.bc_3)}$$

Once the characteristics of the mix have been assessed, it was important to ensure that the RA was correctly mixed. So, it was thought that homogenizing the material by putting it in layers and taking it back in tranches could represent a good manner to homogenize stockpiles.

In the deliverable D1.3, an methodological approach has been proposed and divided in three steps:

- Definition of an optimum way to obtain a representative sample from a stockpile, using a shovel according to the standard EN 932-1, and to compare it to what we can obtain in terms of homogeneity while using a splitter,

- Set up of an experimental program to validate the use of mixing laws when mixing RAs, in order to prevent the stock owner from having to characterize the mix, using the hypothesis that the initial materials were known,

- Validation of a technique to mix stockpiles, consisting in superimposing RAs, in layers, and then taking them back in tranches, with the purpose to obtain similar tranches, each containing approximately the same quantity of initial RAs.

The main conclusions of the investigations as a whole are as follows:

- Sampling by means of a spade in a roof shape stockpile, according to the standard EN 932-1, seems to be an optimum way to make a representative sample. It can be noticed here that locations where RA is sampled by means of a shovel is of a high importance for obtaining a representative sample. This representativeness of a global sample is directly correlated to the samples locations.

- Applying mixing laws for characterizing a mixture of RAs, even for RA containing PmB seems to be reasonable, especially in terms of saving time. As it is often the case, this study however needs more tests for being extrapolated and generalized.

- A first examination of the technique of RA storage in layers has confirmed that this technique was good to obtain similar tranches for re-use.

2.2 Results obtained in Task Group 1.2

RA are complex materials and their use in significant proportions in new asphalts involves a more accurate control of their characteristics. These last ones are essential for asphalt mix design and a key factor for good performances of new asphalt mixtures including a high percentage of RA. Consequently, it is very important to focus on issues that are specifically related to the characterization and technical evaluation of RA and particularly RA containing modified binders for which there is clearly a lack of knowledge and adequate test methods to sample and to analyse them (as shown in the state of the art presented in deliverable D1.1).

One of the topical problem is related with the determination of the binder content for which the current European test methods for extraction and recovery of binder in RA are only normative for RA with pure binders and give only indicative informative for RA containing PmB. A key point for RA containing PmB is to know if the used whole recovery procedure permits to fully extract PmB while modifying as little as possible the properties of PmB. In this study, it has been tried to identify the impact of extraction procedure on the PmB properties. So, as binder extraction and recovery is a essential step to determine the basic characteristics of RA, the impact of the couple testing method/solvent on the binder content and binder characteristics has been assessed in the frame of a comparative study of different standardized methods according to the European test methods EN 12697-1 and EN 12697-3 applied to RA with PmB. The six laboratories involved in task group 1.2 have participated in a round robin test, in which three different bituminous materials have been evaluated:

- One Stone Mastic Asphalt with modified bitumen as virgin binder and including 15% of RA (also with PmB; source not known);
- 2 Reclaimed Asphalts with Polymer modified Bitumens. The polymer SBS (Styrene-Butadiene-Styrene) being the most common polymer modifier used at this time, only RA with elastomer modified bitumens have been selected:
    - One Porous Reclaimed Asphalt that was sampled in 2009 directly after milling from porous asphalt layer of age 10 years; the binder used for this RA is a physical SBS modified bitumen.
    - One Reclaimed Asphalt that was sampled in 2004 directly after milling from porous asphalt layer of age 10 years; the binder used for this RA is a chemically linked SBS modified bitumen.

The results of this round robin test are described in deliverable D1.2. According to the obtained results in this preliminary study, it seems that there is no impact of the couple testing method/solvent for the determination of the soluble binder content on a new asphalt mixture with modified bitumen and including a low content of RA. Problems appear for RA containing aged bitumen with PmB, for which the ageing level of the bitumen in the RA, combined with the presence of polymers leads to a more difficult recovery. At this time, the European standards EN 12697-1 and EN 12697-3 appear to be not clear enough when asphalt contains PmB. The same trends can be observed on the characteristics (namely penetration, softening point, oxidation degree, polymer content, complex modulus, and ductility force) of recovered PmB, the values being more scattered for RA than for new mix, whatever the measured properties.

According to these first results (presented in deliverable D1.2), it was decided to go further in the study of extraction procedures. The aim of this second study was to identify the impact of experimental parameters (solvent, methods and temperature of dissolution) on the determination of soluble binder content of RA containing PmB and on the characteristics of recovered PmB. Consequently, the proposed experimental program described in deliverable D1.4 consisted in producing artificial RA with a defined binder content and known binder properties in laboratory. Then, the different recovery methods were applied according to the standards EN 12697-1 and EN 12697-3. As theses methods describe a large range of methods and solvents that can be chosen to carry out the tests, the different results have been compared to the "true" ones in order to evaluate the adequacy of the methods and solvents proposed in standards to characterize RA with PmB. This experimental program (described in deliverable D1.4) has been performed on two kinds of asphalt mixes : the first one was a "standard mix with unmodified binder" used as reference (called "AC11") and the second one was a PmB mix with "standard" PmB (called "MR"). A part of the study has consisted in aging the mixes and so analyses were performed before and after laboratory ageing. The binder used to produce both the unaged and aged MR mixes was announced as modified with SBS. By FTIR measurements it could be shown that the mix actually contained some other kind of binder, though the exact modifier could not be identified due to lacking reference spectra. Though this observation indicated the possibility to indicate the presence of known and unknown modifiers in a mixture.

Following conclusions have been drawn on all the analyses presented in the deliverable D1.4 (RA prepared in laboratory) but also thanks to results obtained from RA taken on site (described in deliverable D1.2):

- For the determination of soluble binder content, the testing method and the solvent have no impact on a new asphalt mixture with modified bitumen and including a low content of RA. Problems appear when characterizing RA with Polymer modified Bitumen. The more scattered results are obtained for RA with a physical SBS modified bitumen, but the largest span is obtained for RA with chemically linked SBS modified bitumen. From a technical point of view, the recovery of the binder from a RA with PmB is more difficult than for a new asphalt with PmB, and the choice of the testing method and of the solvent seems linked with the type of binder (physically bond or cross-linked PmB) : the impact is more pronounced for RA with a chemically linked SBS modified bitumen maybe due to some solubility problems linked to the nature and temperature of used solvent.

However, the number of participating laboratories and repetitions were limited, so these results need to be confirmed by a similar round robin test at a larger scale, with more participants and more repetitions. It also must be performed on RA with PmB of controlled quality (laboratory-aged).

- Among the different characteristics measured on recovered binders from RA (namely penetration, softening point, oxidation degree, polymer content, complex modulus, and ductility force) two characteristics have been identified as strongly related to the assessment of the end of life of RA, that are:
  - the content of carbonyls (linked to the oxidative ageing)

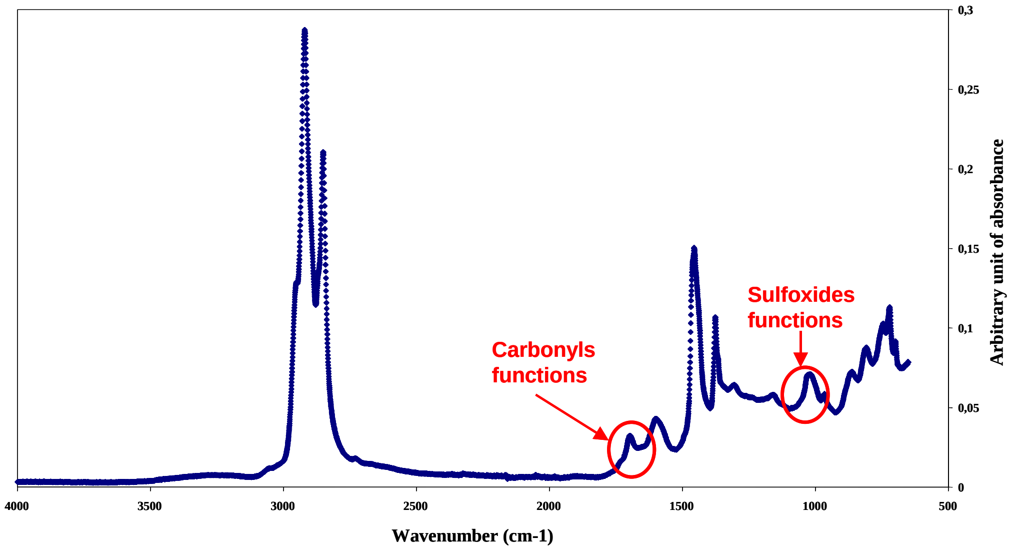

Figure 2: identification of the presence of carbonyls functions based on FTIR spectrum of a recovered bitumen from RA.

  - the complex modulus at 25°C and 52°C (either 1.6 Hz or 10 Hz) (linked to the hardening of binder).

So, these two characteristics could be used for the choice of adding binder as indicators for the recyclability potential of aged binder as described in the final deliverable D2.7 of Work Package 2 for which the main goal was to identify the impact of RA quality and characteristics on performance of asphalt containing RA. In the future, it would be interesting to define the threshold values of these characteristics for recycling a given RA. This can be done only by collecting data.

These results were implemented to the mix design procedure proposed in deliverable D2.7. In case of presence of polymers in the binder extracted from RA it is recommended to ensure new materials performance quality by laboratory performance tests after a theoretical mix design is conducted in order to avoid incompatibilities between RA properties in the new mix.

In addition to the composition of RA binder that is an important issue related to the possible re-use for these materials, it is also necessary to check that the aggregate characteristics, according to the selected proportion of RA, are also appropriate for being reused, into wearing courses or more appropriate road layers, according to the characteristics of this RA. In particular, it may be essential to ensure that its grading does not preclude its recycling into a new asphalt mix, especially when levels of recycling become significant. Checking a RA grading curve is usually performed by sieving. However, this method is time consuming; hence it may not be appropriate to the production rate of mixing plants. To overcome this problem, the use of the VDG40 videograder was suggested since this device is able to rapidly determine the grading curve of aggregates. As it is an alternative optoelectronic device to the classical sieving method, comparative tests with traditional testing means have been performed on different kinds of RA in order to validate the use of this device for such materials.

This study described more in detail in deliverable D1.5 shows that the videograder may be used for quick checking of the homogeneity of RA grading at the mixing plant. Despite the videograder is not appropriate to deliver the actual sieve grading curve of the RA aggregates and especially not the fines content, this apparatus presents a good repeatability in terms of measurement. It can also be used to detect big lumps that could be problematic for the reuse of the RA in a new hot bituminous mix.

2.3 Results obtained in Task Group 1.3

To improve the environmental characterization of reclaimed asphalt (RA), there is a need for laboratory test methods and test scenarios that can support practitioners in assessing emissions due to release of particles, vapours/fumes and leachates, respectively. Consequently, the aim of deliverable D1.6 was to provide guidance on the selection of appropriate test methods for the environmental characterization of air- and water-borne emissions arising from the use of RA. In addition, characterization tests that can support the environmental assessment of RA, such as ecotoxicological and bioavailability tests, have also been included. Towards this end a number of specific objectives were identified. These include:

- Identification of potentially hazardous organic compounds susceptible to leaching from RA

- Evaluation of methods suitable for assessing the potential for leaching of hazardous compounds from RA
- Further characterizsation of this leaching behaviour through appropriate ecotoxicity testing
- Evaluation of a test method for assessment of the bioavailability of organic constituents associated with RA
- Evaluation of an appropriate test method for airborne emissions arising in mixing conditions at manufacturing temperature.

The number of candidate contaminants that can be analysed through environmental testing is vast as seen in the state of the art of deliverable D1.1. Effective characterization of RA requires that potentially hazardous compounds susceptible to leaching from RA are identified. To provide this information a literature review and a screening analysis were performed (described in deliverables D1.1 and D1.6). The literature review focused on metals while the laboratory study (qualitative and semi-quantitative screening analysis) aimed to identify organic compounds. The main aim of this task was to provide a good basis on which the decision of target compounds, to include in the following analysis, could be made. The main conclusion from this preliminary screening analysis is that:

- For the inorganic compounds it was decided to analyse leachates for the following elements: barium (Ba), cadmium (Cd), cobalt (Co), chromium (Cr), cupper (Cu), manganese (Mn), magnesium (Mg), nickel (Ni), lead (Pb), antimony (Sb), vanadium (V) and zinc (Zn).
- For the organic substances, polycyclic aromatic hydrocarbons (PAHs) and benzothiazole (if the tested material contains rubber-asphalt) were identified as the most important organic compounds to collect data for. Alkanes, adipates and phthalates were rejected due to a combination of low concentrations and low toxicity levels.

As PAH was identified as an important group of hazardous organic compounds existing in emissions that arise at different life-cycle stages of RA, it was also decided to quantify the total amount of PAHs in the studied solid RA materials in order to relate the obtained emissions released to the amount in the solid material. At present there are several methodologies available, using different pre-treatment and analytical quantification methods, leading to variation in results obtained depending on the selection of methodology. The key point is that the choice of sample preparation can have a major impact on quantifying PAHs. The results of the here tested RA samples, according to three different experimental methodologies, confirmed that the choices of sample preparation can have a major effect on PAH content and PAH compound distribution. The study also showed that different particle fractions of a RA material can have major differences in their PAH-content. The source of the PAH in the tested RA was confined to a distinct layer of tar asphalt before sample preparation. One possible reason for the difference in the amount of PAH can be that the binder from the tar asphalt layer after crumbling was enriched in the smaller fraction of the RA tested (particles of <1mm) or that the overall binder content was enriched (PAHs are bound to the organic binder phase of the asphalt). However, there was no available data on binder content, or binder quality, in the different fractions tested and hence these assumptions could not be supported by additional data.

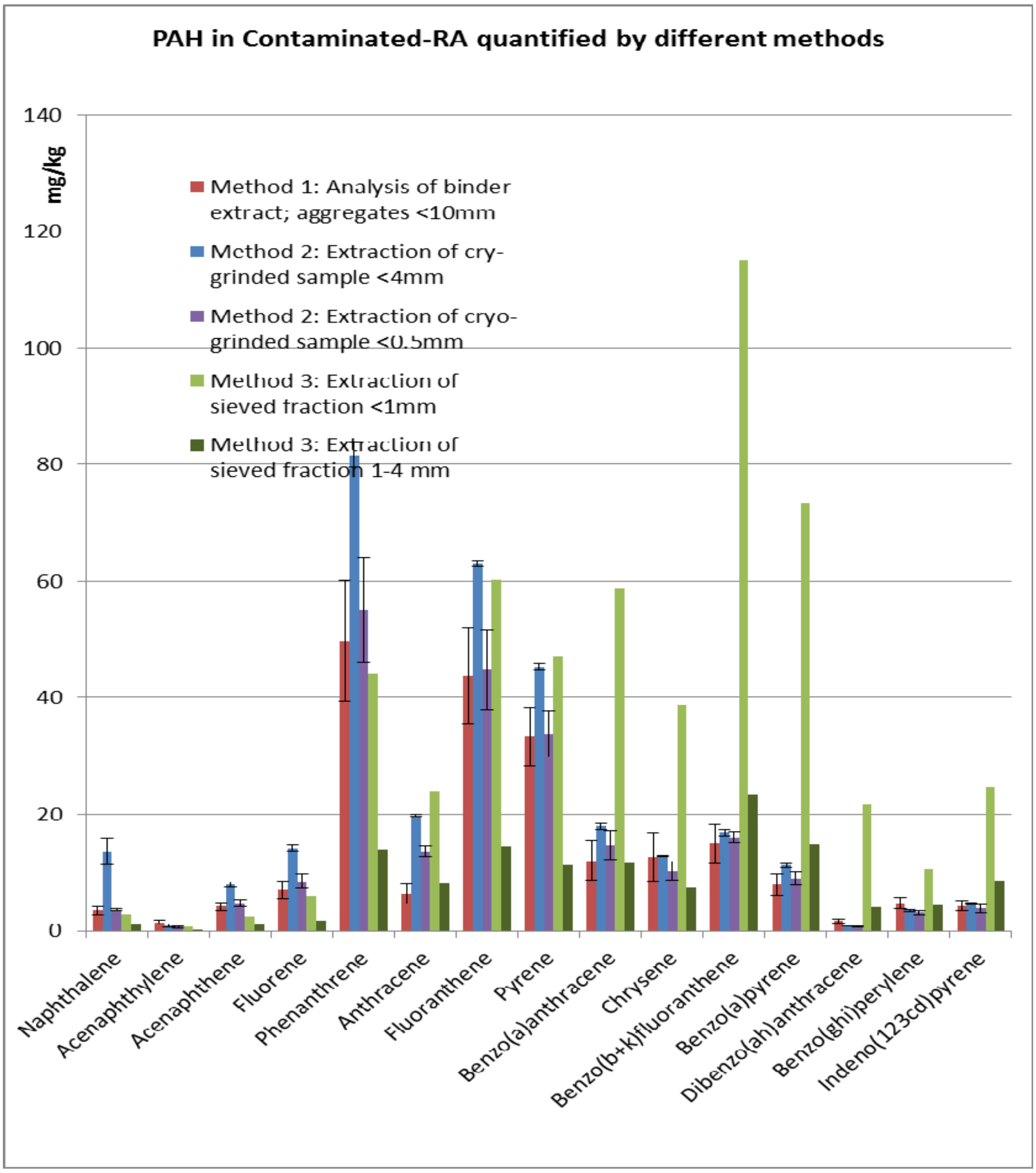

Figure 3: Amounts of PAHs in sub-samples of a reference material ("Contaminated-RA") [mg/kg] quantified with different methods.

Three different leaching methods (a percolation test, a batch test and a recirculated column test), were selected for evaluation and applied on a reference material ("Contaminated-RA") to assess the repeatability of the various methods. Although based on a very limited number of samples the conclusion is that all the tests had a sufficient repeatability for use on RA with a high level of contaminants. The results from a "laboratory comparison" (only two participating laboratories) on the percolation test showed that the experimental design can have a high impact on the leaching results of both HOCs and inorganic compounds. It was also found that the development of a test for simultaneous assessment of both inorganic and organic compounds is very difficult.

Furthermore, in comparison to the other test methods the batch test does not seem suitable for assessing highly hydrophobic organic compounds. The batch test could possibly be used as compliance test to the percolation method when studying PAHs with low- and medium molecular weight (PAH-L and PAH-M), but will overestimate the leaching of PAH with high molecular weight (PAH-H). For this purpose the recirculated column test may be a better choice.

In order to assess and compare different RA-material's ecotoxicity, the batch test was also used to produce leachates for subsequent ecotoxicity testing; this also included an assessment of the leachates chemical characteristics (mainly the leachates' content of PAHs and inorganic elements). The release of metals from the tested RA materials to water occurred in relatively small quantities and levels of pollution among leachates were generally low. Differences between the material leachates (except leachate from the highly contaminated reference material) were small and total PAH concentrations in these leachates were nearly similar. Ecotoxicity tests can, however, bring additional important information. It has been demonstrated in this study that material leachates with the lowest concentrations of analysed metals and PAH may not represent the lowest risk for water ecosystem. A toxic response obtained in tests on leachate with low concentrations of metals and PAH may be due to the prevalence of "unknown" substances (i.e. substances that were not analysed for;, e.g. other PAHs than the 16 common PAHs, degradation products of PAHs, other organic compounds with origin from the binder, rejuvenators, traffic etc.). It can also be a result of a synergy effect caused by two or more substances that alone exists in harmless concentrations but together causes a toxic response in the tested system. Therefore this study has demonstrated the benefits of ecotoxicity tests for the evaluation of possible threats posed by RA to fresh water ecosystems. It is acknowledged that there is some uncertainty with the test results, but it should be noted that ecotoxicity testing gives information that is not available using chemical tests alone.

Another goal of the task group 1.3 was to identify and test a method for the assessment of the bioavailable fraction of organic pollutants in an asphalt material, i.e. the quantity of the substance that can be mobilised from the RA and become available for bio-uptake or biodegradation. Thus, the use of cyclodextrin extraction as a predictor for microbial degradation of PAHs was investigated on two RA samples. However, none of the studied asphalt materials showed true bi-phasic behaviour with high residual amounts of PAH. On the contrary, the results indicated that most of the PAHs are rather easily desorbed with this extraction method and for several compounds the whole fraction of the contaminant was available, i.e. the extracted accumulated amounts with cyclodextrin were equal to the amounts received with conventional exhausted extraction methods. However, it must be noted though, that no correlation of these results with any biological test methods (uptake to earthworms, microbial degradation etc) has been performed, and hence it is not possible to conclude that the accessible fraction reported here would be equal to the true bioavailable fraction.

Finally, the airborne emissions arising in mixing conditions at manufacturing temperature has been assessed thanks to a model experiment using a new fume generation system in laboratory.

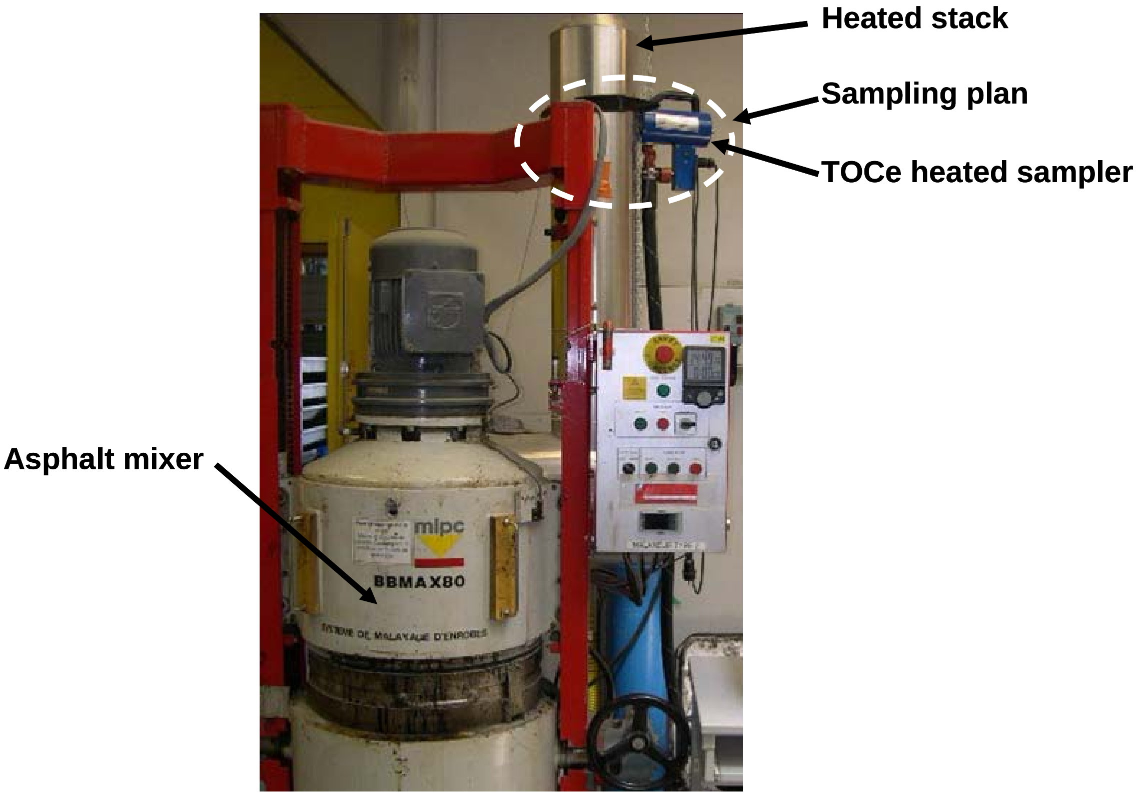

Figure 4: View of IFSTTAR fume generation system (asphalt mixer equipped with an heated stack).

Different indicators (namely TOCe, SVOC, BSM and PAH concentration in fumes) have been used to quantify and rank emissions generated from bituminous mix during mixing at manufacturing temperature. The influence of parameters (RA origin, RA content and binder type) on fume emissions has been assessed. Results show an effect of the binder nature on organic emissions. At same manufacturing temperature (145°C), the use of adding modified binder increases emission level on all studied indicators. For some mixes this effect was found to be lower than the effect of RA origin (and composition). For example, mixes which are composed of the

RA sampled from a stockpile for contaminated asphalt (possibly containing tar) are particularly emissive and generate a high amount of organic compounds compared to the others mixes studied (containing common RA). This laboratory study shows a strong link between bituminous material composition and their emissive potential reported on the three organic parameters studied.

The various test methods described above provide significant insight into the options available in assessing potential environmental impact arising from the use of RA in pavement surface layers. However, for this information to be truly useful for road engineers, it is important that context be provided to the process. This is achieved by combining the results of the various tests with information describing the road construction and the likely pathway that will be taken by contaminants. This will allow engineers to conduct a detailed Risk Assessment of the impact associated with the use of RA in pavement surface courses and to make informed decisions on the best use of this material resource.

3 Proposal of recommendations for a better characterization of RA

3.1 Discussion on sampling of RA

Sampling a RA stockpile for characterization it is not an easy thing to do, since the process is hard to make exactly repeatable. When the material comes from a unique source, like a big jobsite, characterizing RA mainly consists of confirming the homogeneity of the pile. Where mixed RA sources are concerned, our study has shown that mixing laws and handling techniques (layers/tranches) could facilitate future high level use of the material. However, at this step, further tests are required to validate and/or improve their use. When storage of RA is not controlled at all, like in the case of unknown sources, characterizing RA in a safe way seems far more difficult, if not impossible. This does not mean that RA managed in this way cannot be used in high level recycling, but doing so may lead to more risks. According to some stakeholders, the "uncontrolled stockpiles" at asphalt plants mainly consist of asphalts made at the plant (not so uncontrolled) and can be directly reused for making roads around the plant. Developing storage and sampling methods will assist in limiting risks associated with these mixed piles.

In the future, if further sampling is carried out to acquire representative samples and to characterize stockpiles, then the tests of characterization may be further adapted to RA stockpiles. The present protocol for characterizing RA consists of binder extraction and binder characterization and takes considerable time to carry out. Thus defining homogeneity may take too long time, due to the fact that a relevant number of samples have to be characterized. Moreover, making a representative sample needs to identify correctly the location where to plant shovels. This does not represent an easy task. Further efforts should be made to streamline things in the future. Binder content and grading represent the main characteristics of RA in that sense they directly impact the quantity of added materials in the mix. Generally, the Nominal Maximum Aggregate Size (NMAS) determines the utilization potential of the RA but the amount of fines can also have a huge impact on the amount of RA allowed to be introduced in the new mix. It seems also that grading represent an important source of heterogeneity in the material. For this reason it would be interesting to store RA by aggregate size. These two points (changing the method of characterization and storing by aggregate size) may represent interesting studies for the future.

3.2 Discussion on characterization of RA

Characterization of RA is a critical step for asphalt mix design and a key factor to obtain good performances of new asphalt mixtures containing large content of RA. Binder content and grading represent the main characteristics of RA because they directly impact the quantity of added materials in the mix. However, even if the determination of soluble binder content (according to EN 12697-1) does not pose a problem for mixes with pure binders, it is not the case for mixes with PmB. Our results mentioned above clearly indicate that problems appear for RA with PmB, for which the ageing level of the bitumen in the RA, combined with the presence of polymers leads to a more difficult recovery. The European standards EN 12697-1 and EN 12697-3 appear not clear enough when asphalt contains PmB. Moreover, the advanced state of ageing of PmB in reclaimed asphalt may make the extraction process more difficult and not all the standardized methods and solvents available to carry out the test may give similar results. The European standard EN 12697-1 allows different methods for the extraction of the binder. They are to be combined with different methods for separation of the mineral matter. Different solvents have to be used according to differing work safety regulations. Consequently, each partner of task group 1.2 has measured the binder content according to its own methods chosen from EN 12697-1. So, the same samples, representative of the considered RA, are tested by different laboratories, with different methods and different solvents. The observed scattering of the binder content results should be due to extraction methods and solvents as the samples are taken from the same homogenized RA source. In this study, the RA samples were sampled from the same raw material source after homogenization of the material. But, the point is that it was not possible to exhibit the same binder content for same RA using the different methods and solvents allowed in the EN 12697-1. There is clearly an effect of extraction methods and solvents on the binder content. Moreover, due to the important deviation on binder content performed on RA, the laboratory in charge of the analysis must be aware of the limits and the sensitivity of the chosen method. Thus, it has to be verified during the mix design study that mechanical properties of the resulting mix are not impacted by this deviation.

As the results of the reclaimed asphalt characterization are important to estimate the amount of reclaimed asphalt reusable in a new pavement, there is a big interest in considering those questions and to go deeper into the experimental program. It would be interesting to consider only one method with different solvents and only one solvent with different methods, to assess the impact of those parameters on the test. Therefore, the various extraction and recovery methods and solvents could be applied on several kinds of RA samples. As a result it could be established if the recovery method influences the found range of binder content and therefore the allowed content of RA in new hot mix asphalt.

As the composition of RA binder is also an important issue related to the possible re-use of RA and in particular its capacity to mix with the fresh added bitumen by the exchange of viscosities, different characteristics were assessed by laboratory testing after recovery of binder from RA (according to the EN 12697-3). The analysis by Fourier Transform Infrared spectroscopy has revealed firstly that some recovered binders have been contaminated by residual solvent that could artificially modify their characteristics and secondly that binders from MR mixes were not modified by SBS polymer as originally supposed. The infrared spectroscopy is a very promising tool for detecting the presence of polymers and/or pollutants, which cannot be assessed by usual consistency tests. Moreover, for recycling components of a high value (high quality aggregates, modified binders), a key point is to determine the type of bituminous binders (pure or modified) in RA in order to take into account the presence or not of polymers for the choice of adding binder in recycling process.

Among the different characteristics measured on recovered binders from RA (namely penetration, softening point, oxidation degree, polymer content, complex modulus, and ductility force) two characteristics have been identified as strongly related to the assessment of the end of life of RA, that are the content of carbonyls linked to the oxidative ageing and the complex modulus at 25 and 52°C (either 1.6 Hz or 10 Hz) linked to the hardening of binder. As these characteristics could influence the capacity of the RA to mix with the fresh added bitumen by the exchange of viscosities, they can be used as indicators for the recyclability potential of aged binders and for the choice of adding binder. So, in the future, it would be interesting to define the threshold values of these characteristics for recycling a RA. This can be done only by collecting data. However, it has to be noted that these two suggested methods are not very common among European civil engineering laboratories. Moreover, taking infrared spectroscopy and rheological measurements on routine basis could be expensive in manpower and equipment. For those that do not have the possibility to do these tests, the penetration test might give a good preliminary indication. It has to be noted that the Environmental legislation (mass flow limits allowed emitted of chlorinated hydrocarbons from solvent) may even hinder that you are allowed to extract the binder in the first place!

3.3 Discussion on environmental assessment of RA

The testing program has to a large extent been focused on one group of contaminants, namely PAHs. This group of compounds were selected as target substances, based on a screening analysis on organic compounds in leachates from

RA materials. From the screening analysis it was also concluded that benzothiazole should be added to the list of target substances if the tested material contains rubber-asphalt. Alkanes, adipates and phthalates were rejected due to a combination of low concentrations and low toxicity levels.

The screening analysis must not be viewed as a study covering all potential hazardous organic compounds. Only three different RA materials were studied, and focus was put on substances with already well known potential hazardous characteristics. New additives such as e.g. rejuvenators may require screening for other substances and an expansion of the list of target compounds in future test programs for environmental characterization of RA.

The prevalence of PAHs in RA is (together with asbestos) the main environmental factor that restricts reuse. At present, several different laboratory procedures to quantify PAHs in RA are being practised in Europe. This study has tested three different methodologies (including sample preparation, extraction, clean-up procedure and analysis) for quantitative detection of PAHs in RA materials and come to the following conclusions:

- The choices of sample preparation can have a major effect on PAH content and PAH compound distribution. Care is needed in designing the sample preparation methods not to induce substantial bias.
- More studies are needed on how different sample preparation methods can affect the representativeness of laboratory and analytical samples and the distribution of contaminants on different particle size fractions.

In addition, sample preparation (e.g. sieving, drying, crushing and mixing) can also possibly cause cross-contamination in a laboratory and hence sample preparation should be performed separately from other laboratory activities. Furthermore, blanks should always accompany the actual tests. These findings emphasises the need of strict routines at the laboratories when preparing samples for analysis and a clear documentation.

Leaching

The main conclusion from the evaluation of leaching tests for environmental characterization of RA is that the experimental design can have a high impact on the leaching results of both HOCs and inorganic compounds, and that it is complicated to design the test for simultaneous assessment of both inorganic and organic compounds.

Ecotoxicity tests

To be able to perform both conventional chemical analysis (element analysis, PAH-analysis, pH, conductivity, DOC etc.), and ecotoxicity tests on different trophic levels, relatively large volumes of leachtes must be produced for each tested subsample of RA. This limits the choices of suitable leaching methods and at present the most common method for production of eluates for subsequent ecotoxicological testing is the batch leaching test. However, as pointed out above, this test may not produce representative leachates (especially not for highly hydrophobic compounds), nevertheless the batch test was here used to produce leachates for subsequent ecotoxicity testing in order to assess and compare different RA-material's ecotoxicity.

The accompanying chemical analysis showed that differences between the materials leachates (except leachate from the highly contaminated reference material) were small and total PAH concentrations in these leachates were nearly similar. The ecotoxicity tests demonstrated, however, that the RA-leachates with the lowest concentrations of identified metals and PAHs did not represent the lowest risk for a water ecosystem. On the other hand it is true that the leachate could contain "unknown" substances that were not analysed (e.g. other PAHs than the 16 common PAHs, degradation products of PAHs, other organic compounds with origin from the binder, rejuvenators, traffic etc.) and could occur in high concentrations. Therefore this study has demonstrated the benefits of ecotoxicity tests, and that simple usage of chemical analyses for evaluation of possible effects on living organisms could bring misleading information.

Bioaccessibility

The here tested method for assessment of the bioaccessible fraction of organic substances (extraction method with HPCD) does not seem a suitable method to apply to RA material. None of the studied asphalt materials showed true bi-phasic behaviour and near all of the PAHs were rather easily desorbed with this extraction method. The extracted accumulated amount of several of the compounds approached, or was equal to, the amount received with conventional extraction methods for assessment of "total amounts".

The only exceptions are benzo(ghi)perylene and indeno(123cd)pyrene, for which the residual fractions were between 71-88% in the materials after 168h extraction. It cannot, however, be ruled out that also these compounds would have desorbed to a higher degree if higher concentrations of HPCD would have been used (to investigate if the chemical activity is maintained at a maximum).

HPCD is, however, a relatively expensive chemical and if large amounts of HPCD are needed per tested sample it will not be economically feasible to implement this type of method as part of a set of basic environmental test procedures intended to be performed on a regular basis.

It must also be noted that the accessible fraction determined with this method is an operationally defined measure and since we have not correlated our results with any biological test methods (e.g. uptake to earthworms, microbial degradation etc) we cannot conclude that the accessible fraction reported here would be equal to the true bioavailable fraction.

Airborne emissions

The measurements of fumes emissions from different bituminous materials show an effect of the binder nature on the organic emissions. At same manufacturing temperature (145°C), the use of adding modified binder increases emission level on all studied indicators. For some mixes this effect was found to be lower than the effect of RA origin (and composition). For example, mixes which were composed of a highly PAH-contaminated material were particularly emissive and generate a high amount of organic compounds compared to the others mixes studied.

4 Conclusions

In the framework of the WP1 of Re-Road, the requirements for sampling and characterization of RA in laboratory were elaborated with the objective to increase the RA content of surface asphalt mixtures. Based on the results as discussed in the main deliverables D1.2, D1.3, D1.4, D1.5 and D1.6, testing methodologies have been proposed which enable a better characterization of RA in physical, chemical and environmental domains.

Firstly, a sampling method was set up to investigate mixing of known sources of RA. Experiments showed that the mixing laws can be applied in order to characterize a mixture of RAs, including those containing PmB. This approach is potentially advantageous from the perspective of saving time and avoiding excessive testing. This technique, however, requires further investigation to be fully validated and generalized to all possible types of RA.

Then, secondly, the characterization of RA in laboratory has permitted to determine that the recovery of the binder from a RA with PmB is more difficult than for new asphalt with PmB, and the choice of the testing method and the solvent seems linked with the type of modified binder (physically bond or cross-linked PmB). The European norms dealing with this issue are not clear enough and have to be clarified in the future. However, despite this fact, two characteristics of RA binder has been identified as strongly related to the assessment of the end of life of RA, that are the content of carbonyls linked to the oxidative ageing and the complex modulus at 25 and 52°C (either 1.6 Hz or 10 Hz) linked to the hardening of binder. As these characteristics could influence the capacity of the RA to mix with the fresh added bitumen by the exchange of viscosities, they can be used as indicators for the recyclability potential of aged binders and for the choice of adding binder. These results were implemented to the mix design procedure proposed in final deliverable D2.7 of Work Package 2. In case of presence of polymers in the binder extracted from RA it is recommended to ensure new materials performance quality by laboratory performance tests after a theoretical mix design is conducted in order to avoid incompatibilities between RA properties in the new mix.

Finally, to improve the environmental characterization of reclaimed asphalt, different laboratory test methods that can support practitioners in assessing emissions due to release of particles, vapours/fumes and leachates, have been assessed. These various test methods provide significant insight into the options available in assessing potential environmental impact arising from the use of RA in pavement surface layers. However, for this information to be truly useful for road engineers, it is important that context be provided to the process. This is achieved by combining the results of the various tests with information describing the road construction and the likely pathway that will be taken by contaminants (described in details and discussed in Re-Road deliverable 3.3 (McNally et al., 2012) and summarised in deliverable 3.5 (Enell et al., 2012)). This will allow engineers to conduct a detailed Risk Assessment of the impact associated with the use of RA in pavement surface courses and to make informed decisions on the best use of this material resource.

5 References

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THE RE-ROAD PROJECT aims to develop knowledge and innovative technologies for enhanced end of life strategies for asphalt road infrastructures. Such a strategy has an important impact on the energy efficiency and the environmental footprint of the European transport system and fits within the life-cycle thinking which is being introduced in waste policy at European level. It leads to reduction of the need for new raw materials, prevents the creation of waste and the occupation of landfills and consequently minimizes the need to transport these materials to and from the work site and hence reducing energy, pollution including CO_2-emissions.

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EUROPEAN COMMISSION DG RESEARCH
A FP7 Collaborative Project
WORK PROGRAMME Sustainable Surface Transport
SST.2007.1.2.2 End of life strategies for vehicles/vessels and infrastructures

WORK PACKAGES

WP 1 Sampling and Characterization of RA
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WP 2 Impact of RA quality and characteristics on mix design and performance of asphalt containing RA
Konrad Mollenhauer

WP 3 Environmental performance of RA
Anja Enell

WP 4 RA processing and RA management at the mixing plant
Erik Nielsen

WP 5 Performance modelling of RA
Sabine Werkmeister

RA = Reclaimed asphalt