

**- Scientific Report -**

ESF Exploratory Workshop on

**Exploring The Interactions Between  
Carbon And Organic Chemical  
Cycling In Terrestrial Ecosystems**

Lancaster (United Kingdom), 24-26 June 2009

**Convened by:**

**Luca Nizzetto, Andrew Jarvis and Kevin Jones**

# 1. Executive summary

Organic chemical pollution is one of the human products that most dramatically marked the environment during the last century. Industrial processes, modern agriculture, inappropriate usage and wasting processes have led to the global distribution of many chemicals, potentially dangerous for human and ecosystem health. Organic pollutants constitute a major concern given their affinity for the biota and the organic matrices in general. Studies have shown the potential of many organic pollutants to produce unexpected toxic effects in time and places distant from the sources. In particular, persistent semivolatile organic chemicals (SVOCs) undergo long range atmospheric transport following volatilization from source points (1, 2). Among them, the classical persistent organic pollutants (POPs) are the most notorious. Examples are the polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs) such as the DDT and Hexachlorobenzene (HCB) and other byproducts of industrial processes or combustions such as dioxins and furans (PCDD/Fs), which were produced and emitted in the environment during the last decades at a rate of kilotonnes per year (2). Today, compounds with similar characteristics (such as the polybrominated diphenyl ethers, PBDE and other fluorinated compounds, PFOs) are facing the market and represent potential risk for the environment and new challenges for the maintenance of its quality.

Semivolatile and persistent organic compounds are detectable everywhere in earth in abiotic and biotic matrices (3). Some OCPs and PCBs were demonstrated to produce toxic effects in organisms. Hormone disrupting activity has been shown to occur in many fish populations and in organisms at the top of the trophic chains. POPs accumulate in fat tissues, breast milk and genital organs of many target species, including humans. Toxic effects include: cancer, nervous system damages, reproductive and immune system impairments, and hormonal imbalances (4-7).

These findings constituted a serious concern among the scientific community, society and policy makers. International measures have been undertaken in order to mitigate the risk associated to the environmental occurrence of the old stock of pollutants and to prevent new potentially dangerous chemicals to perpetuate the risk. The most important among them is the *2001 convention on persistent organic pollutants* (Stockholm convention) (8). The need to carry on research on the risk associated to the environmental occurrence of these compounds is pressing and essential in order to develop better regulatory tools which meet the necessity of protecting environment and human health and the need of chemicals of modern societies. This concept has been firmly underlined in the Agenda XXI (9), the UN document listing the action and measures that the world governments should undertake in order to reach and maintain a sustainable development model.

A deep knowledge of the behavior of organic chemicals in the environment is a fundamental step to perform the risk assessment and to develop adequate management protocols and regulatory policies. As mentioned above, many large production volume chemicals, such as the classical POPs and the new potential POPs, undergo long range atmospheric transport, a process controlled by continuous temperature dependent condensation/volatilization processes (10, 11). Partitioning between the transport media (air and water) and surfaces is a key factor controlling the global distribution of these pollutants. Organic matrices, such as organic soils, vegetation and biota in general, showed great affinity in accumulating organic pollutants, given their lipophilicity (11-13). For example, it was estimated that 21,000 tonnes of PCBs produced and emitted until the early 70s are now present in background organic surface soils (0–5 cm) globally (12, 13). In organic matrices the enrichment factors defined as the ratio between the concentration in the matrix and the surrounding fluid, can range between  $10^5$  and  $> 10^8$  (14). Organic carbon (C) rich compartments constitute a strong controlling factor determining the environmental fate of those pollutants.

Following this evidence it is reasonable to hypothesize that the dynamic of organic matter turn-over strongly influences the environmental transport of many organic pollutants. Some

studies have shown that particularly productive ecosystems can enhance the deposition of airborne organic pollutants. Deposition of some POPs into forest ecosystems is 3 to 5 times higher than in bare soils (15-19). The accumulation of organic pollutants on primary producers is also a fundamental step in addressing them to the food webs and in therefore determining the exposure of wild life and human populations (20).

Organic matter synthesis and degradation processes dynamically control the stock and the properties of the organic C stored in the different environmental compartments. These processes can play a key role in influencing the accumulation capacity and the partitioning of organic pollutants at global scales. Despite the centrality of the issue, its holistic character, and the implication at the global scales, few studies have directly addressed these points. The C cycle is intimately related to the climate. Global climate change can sensitively influence the balance of production and respiration in the ecosystem and can potentially modify the global balance of SOCs, possibly re-mobilizing old stocks, currently stored in global soil organic matter.

Knowledge about the coupling of organic matter turn-over and the environmental fate of organic pollutants is, at the moment, gravely insufficient. Further research is needed to fill all these gaps and to provide new tools for the risk assessment. The approach to be is, by nature, multidisciplinary. An integration of knowledge coming from biology, ecology and environmental chemistry is strictly required.

The ESF exploratory workshop was aimed to promote cross-fertilization between the fields of ecosystem carbon exchange and environmental organic pollutant fate in order to precipitate rapid advances in conceptual frameworks, monitoring and modelling technology in the organic pollutant fate arena.

This was achieved through the discussion among experts of different aspects of chemical pollution and ecology with the final objective of opening new scenarios for multidisciplinary environmental research.

The workshop took place at Lancaster Environment Centre (LEC) on the 24th-26th June 2009. A total of 17 researchers from 8 European countries and an invited researcher from United States took part to the discussion. Altogether the participants provided the multidisciplinary environment needed to achieve the workshop goals. Included disciplines were: pollutant fate and exposure, chemical modelling, ecology, ecosystem exchange, micrometeorology, engineering and soil ecology.

## **2. Scientific content**

During the first day of the workshop 4 keynote lectures took place. They were aimed to make the multidisciplinary audience familiarizing with basic concepts and state of the art in the different disciplines.

Kevin C. Jones (LEC, UK) presented a comprehensive review on semivolatile persistent organic pollutants. In the presentation he summarized the current knowledge about emission, distribution, and environmental fate of these pollutants. Particular focus was addressed on the role of the environmental pools of organic carbon as reservoirs for SVOCs.

Anders Lindroth (LUCI, Sweden) provided an overview about organic carbon pools in the global environment with particular focus on the relationship with climatic and land-use drivers.

Bernard Ludwig (Kassel University) spoke about the processes driving the fate of soil organic matter (SOM). He introduced key definitions and reviewed the state of the art of the knowledge about SOM fate and turn over.

Finally, Luitgard Schwendenmann, presented a lecture about OC pools in tropical environment.

During the second day, the discussion was organized in 3 sections:

- 1) above ground organic carbon dynamics, and air/surface exchange of pollutants;
- 2) below ground carbon cycle and pollutant processing;
- 3) ecosystem exchange of pollutants: Data sourcing, analytical tools and modelling.

Each section started with two key relevant applications/studies which served as catalysers for the discussion.

In section 1, Luca Nizzetto (LEC) presented an example of current approach used to estimate the net deposition fluxes of SVOCs over a growing forest canopy. Such indirect approach is based on the observation of SVOC concentration change in foliage along with time. Although this study represents one of the few examples of experimental assessment of seasonality in air/surface exchange of SVOCs it has the limit of providing flux estimates at low time resolution and with a considerable uncertainty.

In the following presentation, Judith Perlinger (Michigan Univeristy of Technology) presented a novel method for the direct air-surface exchange flux measurement, based on micrometeorological approaches and on the use of multicapillary collection denuders as atmospheric samplers for SVOCs. Dr Perlinguer achievements currently represent the most advanced attempts of addressing air-surface exchange of SVOCs.

In section 2, Lukas Wick introduced his studies about the role of fungi hyphae in enhancing availability of SVOCs to degrading micro-organisms. Fungi hyphae serve as vectors increasing the mobility of micro-organisms and providing them a quicker access to the contaminant associated to SOM and not readily available.

In the second talk, Donald Monteith (ECH, UK) presented a review of his long term monitoring of DOC release from catchments. During the last decade a significant increase of DOC concentration on river has been observed across northern Europe. It has been postulated that such increase is a consequence of increased release from catchment soils triggered by changed properties in atmospheric depositions. Some author suggested climate change as one of the potential causes.

The observed phenomenon constitute a potential vector driving to the release of old burden of pollutant stored in soil.

In section 3, Ana Cabrerizo and Jordi Dachs summarized their results about measuring concentration gradients of SVOCs over soils at small spatial scales. The adopted sampling technique allowed significant gradients to be observed for selected compounds. Such result is promising in terms of possibility of measuring deposition or volatilization fluxes of SVOCs, although, at this stage no attempt was performed in order to inform the measurements with micrometeorological parameters and achieve flux estimates. These results constitute an important step forward suggesting that a combined study of air-soil C exchange and SVOC exchange can potentially be performed at the small scale, providing the conditions for a direct investigation of the relationship between C and pollutant cycles.

In the same section, Davide Ghirardello and Antonio Di Guardo presented a computer model aimed to simulate the long term fate of SVOCs in a dynamic soil. Although the model considered, among other processes, the mass balance of SOM and soil water and provided a level of dynamism never attempted before, the simulation results were not satisfactory when checked against experimental data.

During the last day, the participants were asked to discuss about relevant issues emerged during the sections and explore possibilities for future joint research/initiatives.

In particular, large space was dedicated to the discussion about the possibility of carrying on networking initiatives within the ESF scheme underlying, as a core idea, the promotion of actions toward the development of new integrated assessment methods for chemical environmental fate and risk assessment.

### **3. Assessment of the results, contribution to the future direction of the field**

We report here the relevant points emerged from the discussion.

#### **1) Needs of tools for the direct measurement of pollutant air-surface exchange flux.**

The discussion focused on the semivolatile organic pollutants (SVOCs), given their high affinity for organic carbon, their relevance (in terms of size of the environmental burden and toxicity), and the still few information about the interconnection between their environmental fate and the carbon cycle.

Our comprehension of the interactions between pollutant fate, C cycle and physical environment is indeed limited by the difficulty of performing (through the traditional analytical and sampling methods), direct observations on the inter-compartment exchange fluxes and in particular of the air-surface exchange. After more than 30 years of development in SVOC analytical chemistry, our comprehension of the mechanisms controlling the environmental fate is still merely anchored to the measurement of concentrations. In the meanwhile, in many other fields (C ecosystem exchange, volatile organic compounds etc.) the application of micrometeorological techniques and the availability of highly sensitive detection devices, allowed air-surface exchange fluxes of C, heat, water vapour and volatile compounds, to be measured directly, with huge benefits for the advance of the knowledge in the respective fields.

This situation forced “SVOC” researchers in developing a knowledge of pollutant fate mainly built on model simulations. Researchers have been creating regional or global scale models aimed to theoretically predict inter-compartment exchange (and in particular air-surface exchange). Such models were calibrated and “validated” against concentration data.

Such framework however is affected by major limits:

- Although fundamental to assess environmental exposure, concentration data are “integrated” parameters of non-linear processes, and are therefore complicated to handle or inadequate when used to test models or assess dynamic processes.
- There currently is no mean to assess if the experimental concentration data represent a steady state scenario or not. Despite this, when used for assessing mechanistic fate models, concentration data are often assumed to represent steady state conditions, while often such assumption can not be verified experimentally.

These aspects posed a serious limit in our comprehension of the processes controlling the air-surface exchange of SVOCs. As a consequence of this, extrapolations of not fully understood and/or characterized processes (at the small scale), has been carried on, through modelling, at growing spatial scales, driving to conclusions often unverifiable.

The key technological step required to fill these gaps is to move from a process assessment based solely on ‘concentration data’ to one based on both concentration and flux data.

Micrometeorological techniques jointly to recent achievements in SVOC analytical chemistry, which allow concentration measurements to be carried on a more time/cost effective way (i.e. using multicapillary collection denuders), seems to offer the chance of overtaking current limitations.

The application of this or similar techniques for assessing the air-surface exchange of SVOCs over vegetated system (i.e. forests, agricultural fields, soils etc.) will potentially provide the

required tool to investigate the coupling between C cycle and SVOC cycling and will allow more focused and detailed studies to be carried out for mechanism comprehension and model building.

An additional important advantage offered by the micrometeorological applications, is the possibility of performing observations at different spatial scales (for example by performing observations at different heights on a tower) and therefore assessing the relative importance of the different driving processes at the different scales.

From the discussion it clearly emerged that the required technology for implementing SVOC Micromet informed flux measurements is potentially already available, and to make it truly operative, a multidisciplinary joint action should be carried on by ecologists, environmental chemists and engineers. It was discussed that gradient based methods or relaxed eddy accumulation methods employed under quasi-stable boundary layer conditions were appropriate. There were significant discussions about the limits of the sample frequency for POP/SVOCs measurements and it was felt that sub 4 hours was readily achievable. The group moved on to discuss the Bowen ratio methods Judith Perlinger had been developing. Again, given the relative inexpense of sonic anemometers it was felt that deriving eddy diffusivities from sensible heat was the way forward. Covariance methods were clearly out of reach, but the group felt that disjunct methods were worth further consideration, although this required resolving an automated sampling cartridge, which it became obvious, would become the most expensive component of the system.

## **2) Fate of pollutants in soils and ecological processes**

The global distribution of many persistent organic pollutants in terrestrial environment is controlled by SOM distribution. The direct relationship between pollutant concentration and SOM content has been historically exploited by models to predict fate of pollutants at different spatial scales.

Unfortunately current comprehension on the mechanism controlling the partitioning of organic pollutants on the SOM fails in addressing the role of dynamic ecological drivers which affect the short and long term fate of SOM in soils.

SOM consists mainly of stabilized organic matter (humus) and litter (plant residues). The traditional subdivision of humus into fulvic acids, humic acids and humins is not generally regarded as useful. Stocks of SOM vary widely and average stocks in the soil are in the range of 42 t C ha<sup>-1</sup> (tropical grasslands) to 723 t C ha<sup>-1</sup> (swamps and marshes). Litter consists of a variety of compounds. Important components are cellulose, hemicellulose, lignin, proteins, lipids, starch and monomeric sugars. A typical temporal course of the mineralisation and humification of plant residues consists of a fast mineralization of especially cellulose, starch and monomeric sugars, whereas the decay and transformation of lignin is much slower. Humic substances cover a wide range of molecular weights (ranging from 2000 to more than 300 000 g mol<sup>-1</sup>), carbon (45 to 62 %) and oxygen contents (48 to 30 %) and exchange acidity. Driving forces of SOM dynamics are manifold and SOM stocks and composition are mainly affected by climate, by land use including soil management (e.g., tillage vs. no-tillage) and by the quality and quantity of carbon input.

Current attempts of describing the fate of SVOCs in soils still miss-consider the fundamental ecological processes and the complexity described here. Our comprehension on the relationship between SOM and organic persistent pollutants do not take yet into consideration the knowledge available from soil ecology. In order to overtake these limits, a joint effort is required, where already available methods and conceptual frameworks adopted in soil ecology could be exploited for the understanding of SVOC fate.

### **3) Biodegradation in soil and bioavailability**

Degradation of SVOCs in soils has been mainly discussed in terms of bio-availability to degrading micro-organisms. Bio-availability appears to be one of the key drivers controlling the kinetics of the degradation of SVOCs in soil. A novel approach presented here by Lukas Wick focus on the role of fungi hyphae as vectors for micro-organisms mobilization and chemical transport through the soil. The developed experimental approach, performed under controlled conditions, allow a fine analysis of the system providing the conditions to isolate variables and effectively measure key parameters such as transport kinetics of micro-organisms and degradation rates. Lukas Wick's studies appear to represent a promising platform to develop new conceptual frameworks which in future will hopefully be able to provide a finer mechanistic description of the overall degradation of SVOCs in soils.

The comprehension of the mechanisms controlling the degradation of SVOCs in soils is fundamental to improve the effectiveness of bio-remediation tools. If on one side, in recent years, a wide literature became available about selection/engineering of SVOC biodegrading micro-organisms, aspects concerning the interactions between these organisms and the soil ecosystem did not receive sufficient attention. Once more, the discussion underlined the need of adopting a more ecological approach in process assessment.

### **4) Soil dissolved organic matter export and fate of SVOCs.**

Productive ecosystems, such as boreal catchments, enhance deposition of airborne SVOCs, given the large availability of OC. Accumulation on forest canopy and consequent transfer to soil through litter deposition plays a significant role for the accumulation of these pollutants in soils (16).

Within this scenario, the importance of DOC-associated transport of SVOCs between the terrestrial and aquatic system has not yet been sufficiently investigated, although this could represent a relevant pathway determining fresh water biota exposure to organic pollutants.

One of the key questions currently addressed in ecological research is how the carbon cycle of forested ecosystems will be affected under global change. Changes in climate (temperature, precipitation amount and intensity) may in concert affect vegetation and soils processes so that central carbon fluxes (photosynthesis, litter production, soil organic matter mineralization, lateral transport from soils to surface waters) will be altered, resulting in a changed terrestrial carbon sink strength. Changed hydrology usually leads to concomitant changes in lateral DOC transport from catchment to surface waters, due to more superficial run-off and higher DOC concentrations (21) and/or a simple increase in run-off. DOC concentrations in freshwaters have increased significantly (from 14 to 22% in Norway (22) and up to 95% in the UK (23) in boreal ecosystems in the past decades, possibly as a response to reduced acid deposition (24) and changes in precipitation patterns.

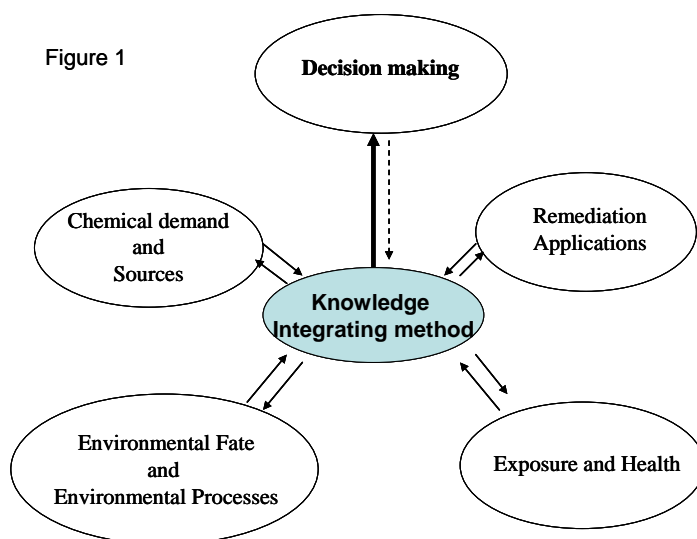
The described phenomena could affect the large burden of "old" POPs stored in boreal soils, potentially favouring its re-mobilization and in particular the transport toward the aquatic ecosystem. This will potentially have implications for the exposure of the biota which could receive enhanced loads of POPs in the short-medium term. Additionally, climate change may affect global distribution and transport of POPs in the medium and longer term through changes in air circulation

Few studies, so far have directly addressed the importance of carbon and water-driven release of SVOCs, their seasonality and the direct implications for aquatic environment in a holistic and multidisciplinary way. Current knowledge does not allow a proper evaluation of these issues.

## 5) Implementing methods for the integrated risk assessment of Chemicals

The discussion carried on during the last part of the workshop, overtook the boundaries of the core topic on which the meeting was based in order to include the issues described here in a wider perspective. This discussion was carried on under the perspective of implementing future networking actions.

The study on environmental fate and exposure to SVOCs (and therefore on the interaction between SVOCs and biogeochemical cycle) represent a component of a wider knowledge set up that, all together, constitutes the platform on which decision making on chemical management is based.



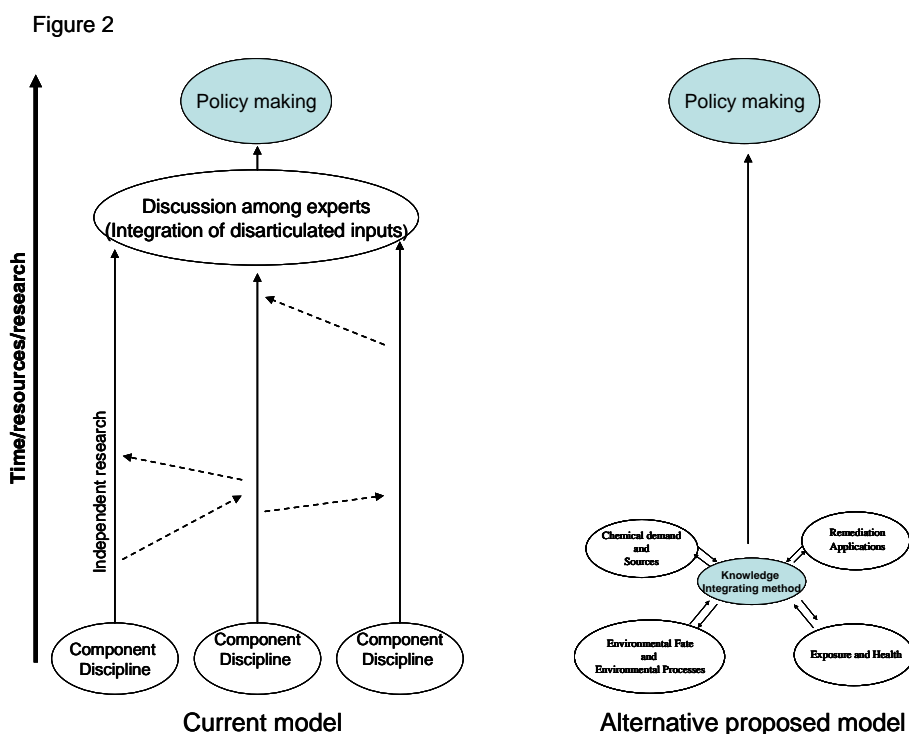
During the discussion “chemical management” was defined as constituted by the component area represented in figure 1, in which the issues discussed here are part of the “Environmental fate and Environmental Processes” component.

As we can see, policies regarding chemicals are the results of complex multidiscipline - multi parameter inputs. However the integration of such inputs is only performed at high level (close to the “end side” of the production line driving to the decision making) and is often based on qualitative or semi-quantitative approaches (discussion among experts) given that the different component areas provide “disarticulated inputs”.

This process is not fully scientifically sounding and the uncertainty range around the information delivered to the policy maker still give potential space to the influence of lobbying and ideologies.

Two models (the current one and the proposed one) for the decision making production line are schematized in figure 2.

The figure shows how, in the current model, multidisciplinary research is inhibited and only associated to few independent / exploratory initiatives (dotted arrows). Additionally, resources dedicated to the integration of knowledge are limited, giving space mainly to qualitative approaches performed at the end of the production line.





In the proposed model the integration of component areas occurs at the beginning, and knowledge integration itself arises to be a discipline, aimed to develop quantitative methods to aggregate inputs from the different components area. In this way most of the resources are addressed to deliver an integrated assessment more than disarticulated inputs. Additionally all the component areas can receive “guidelines” from the “knowledge integrating method” in order to focus the independent research toward the delivery of outcomes which can be quantitatively included in the integration.

The focus of the future networking initiatives will be the development of quantitative “Knowledge Integrating Methods” to serve as “glue” for the different component areas and to promote the implementation of the new model applied to chemical management. In synthesis: to develop tools to populate the central area in figure 1.

## 4. Final programme

### Wednesday 24 June 2009

<i>morning</i>	<i>Arrival</i>
13.30-14.10	<i>Welcome and buffet</i>
14.10-14.20	<b>Introduction to the Workshop</b> <b>Luca Nizzetto</b> (Lancaster University, UK)
14.20-14.40	<b>Presentation of the European Science Foundation (ESF)</b> <b>Aslian Kerç</b> (Standing Committee for Physical and Engineering Sciences - PESC / Standing Committee for Life, Earth and Environmental Sciences - LESC)
14.40-15.40	<b>Participant introductions</b>
15.40-16.00	<b>Keynote lecture: “Global occurrence of persistent organic chemicals: the role of organic matter”</b> <b>Kevin Jones</b> (Lancaster University, UK)
16.00-16.20	<i>Coffee / Tea Break</i>
16.20-16.50	<b>Keynote lecture: “Organic carbon stocks in different terrestrial ecosystems: metrics, dynamics, driving forces”</b> <b>Anders Lindroth</b> (Lund University, SE)
16.50-17.20	<b>Keynote lecture: “Organic matter in soils: characteristics, metrics, dynamics, driving forces”</b> <b>Bernard Ludwig</b> (University of Kassel, DE)
17.20-17.40	<b>Keynote lecture: “The determinants of carbon pools and fluxes in tropical ecosystems”</b> <b>Luitgard Schwedenmann</b> (Burckhard Institute of Tropical Silviculture, DE)
17.40–17.50	<b>Opportunities and outcomes from the workshop</b> <b>Luca Nizzetto</b> (Lancaster University, UK)
17:50-18.00	<b>Carbon cycle and environmental cycling of pollutants: “Keynotes”</b> <b>Andrew Jarvis</b> (Lancaster University, UK)

## Thursday 25 June 2009

- 08.30-10.20      **Section: Above ground organic carbon dynamics, and air/surface exchange of pollutants**  
Facilitators: **Judith Perlinger** (Michigan Technological University, USA)  
**Luca Nizzetto** (Lancaster University, UK)  
Rapporteur: **Andrew Jarvis** (Lancaster University, UK)
- 10.20-10.40      *Coffee / Tea Break*
- 10.40-12.30      **Section: Below ground carbon cycle and pollutant processing**  
Facilitators: **Lukas Wick** (Helmholtz Centre for Environmental Research, DE)  
**Donald Monteith** (Centre for Ecology and Hydrology, UK)  
Rapporteur: **Kyrk Semple** (Lancaster University, UK)
- 12.30-14.00      *Lunch*
- 14.00-15.50      **Section: ecosystem exchange of pollutants: Data sourcing, analytical tools and modelling**  
Facilitators: **Jordi Dachs** (IIQAB-CSIC, CA)  
**Antonio Di Guardo** (University of Insubria, IT)  
Rapporteur: **Matthew MacLeod** (ETH, SWI)  
**Kay Hansen** (NERI, DN)
- 15.50-16.10      *Coffee / tea break*
- 16.10-16.40      **Rapporteur's summary**
- 16.40-17.30      **Discussion: identification of topic areas and research needs.**  
Definition of work groups
- 19.30              *Social dinner in the evening (restaurant "Quite Simply French" in Lancaster)*

## Friday 26 June 2009

- 08.30-11.30      **Work group discussion**
- 11.30-11.50      *Coffee / Tea Break*
- 11.50-12.20      **Rapporteur's summary from work group discussion**
- 12.20-13.30      **What's next**
- 13.30              *Buffet lunch and Departure*

## 5. Statistical information on participants

Total participant number	17
Number of participant younger than 35	5 (29%)
Number of female participants	4 (24%)
Number of participating countries	9
from UK	5 (29%)
from Germany	3 (17%)
from Italy	2 (12%)
from Spain	2
from Denmark	1 (6%)
from Sweden	1
from Switzerland	1
from Turkey	1
from USA	1
Number of PhD students	2 (12%)

## 6. List of participants

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