Convocatorias 2017
Proyectos EXCELENCIA y Proyectos RETOS
AGENCIA ESTATAL DE INVESTIGACIÓN

AVISO IMPORTANTE

En virtud del artículo 16 de la convocatoria **NO SE ACEPTARÁN NI SERÁN SUBSANABLES MEMORIAS CIENTÍFICO-TÉCNICAS** que no se presenten en este formato.

Es obligatorio que la memoria contenga los tres apartados (A, B y C). La parte C de la memoria no podrá exceder de 20 páginas.

Lea detenidamente las instrucciones para rellenar correctamente esta memoria, disponibles en la web de la convocatoria.

Parte A: RESUMEN DE LA PROPUESTA/SUMMARY OF THE PROPOSAL

INVESTIGADOR PRINCIPAL 1 (Nombre y apellidos)**:**

Oriol Jorba Casellas

INVESTIGADOR PRINCIPAL 2 (Nombre y apellidos)**:**

TÍTULO DEL PROYECTO: Química del carbón marrón: modelización de la absorción de amoníaco por los aerosoles orgánicos secundarios y su efecto en el forzamiento radiativo

ACRÓNIMO: BROWNING

RESUMEN Máximo 3500 caracteres (incluyendo espacios en blanco):

Los aerosoles orgánicos (AO) afectan al clima y a la salud humana, pero aún existe una elevada incertidumbre en sus emisiones y evolución. Los AO representan una fracción importante del material particulado fino, 20 al 90%. Estudios recientes han mostrado que los AO secundarios (SOA) representan también una fracción importante del AO total. Los AO reflejan eficientemente la radiación visible y contribuye al enfriamiento de la atmosfera. Sin embargo, una fracción significativa del AO también absorbe radiación en el visible y UV-cercano. Esta fracción es conocida con el nombre de "Carbón Marrón" (CM). El CM contribuye junto al Carbón Negro y el aerosol mineral al calentamiento de la atmosfera. Estudios recientes han identificado que determinado SOA con propiedades de reflexión dominante sufre un cambio de color al estar expuesto a concentraciones de componentes con contenido de nitrógeno reducido, como es el amoníaco, el amonio derivado del sulfato-amónico o otro tipo de sales y amino ácidos. La contribución de este AO con propiedades de absorción al calentamiento global del CM es aún muy incierto y puede representar una contribución importante en zonas donde la emisión de amoníaco o sales de amonio sean elevadas.

El objetivo principal de esta propuesta es el estudio del efecto de la reacción de especies con contenido de nitrógeno con los aerosoles SOA en las propiedades ópticas y la concentración final de SOA antropogénico y biogénico empleando técnicas de medida y de modelización. Proponemos implementar una nueva química del amoníaco con SOA en un modelo atmosférico-químico, el NMMB-MONARCHv1.0, para cuantificar la fracción de CM de origen primario y secundario, y la contribución de este SOA absorbente al forzamiento radiativo del AO. También proponemos realizar una campaña experimental en campo abierto en el noreste de España para estudiar la relevancia de esta nueva química en una

región caracterizada por concentraciones de amoníaco y ozono elevadas. Se plantea realizar medidas de concentración en superficie de amoníaco, ozono y composición química de los aerosoles, junto con medidas de propiedades ópticas (absorción y reflexión). Estos datos representarán una fuente única de información sobre la contribución del CM secundario en el noreste de España.

PALABRAS CLAVE: Química atmosférica, aerosoles orgánicos, amoníaco, absorción, modelización

TITLE OF THE PROJECT: Brown carbon chemistry: modeling the uptake of ammonia on secondary organic aerosols and its effect on the radiative forcing

ACRONYM: BROWNING

SUMMARY Maximum 3500 characters (including spaces):

Organic aerosols (OA) affect climate forcing and human health, but still large uncertainties exist in their sources and evolution. OA makes up a large fraction of the submicron particulate matter, 20 to 90%. Recent studies have shown that secondary organic aerosols (SOA) accounts for a large fraction of the OA burden. Most OA efficiently scatter visible radiation and contribute to a cooling effect in the atmosphere. However, a significant fraction of OA absorbs radiation in the near-UV and visible ranges. Such fraction is known as "Brown Carbon" (BrC). BrC contributes together with Black Carbon and mineral dust to warming the atmosphere. Recent studies have identified that some initially scattering SOA undergoes browning during exposure to reduced nitrogen compounds such as ammonia, ammonium from dissolved ammonium sulfate or other salts, and amino acids. The contribution of such light-absorbing OA to the total warming effect of BrC is still highly uncertain and it may be significant in areas that have high emissions of ammonia or dissolved ammonium salts.

The main objective of this proposal is to explore the effect of the reactive uptake of nitrogen-containing species by SOA on the optical properties and burden of anthropogenic and biogenic SOA using both experimental and modeling approaches. We propose implementing new NH3-SOA chemistry in a state-of-the-art atmospheric-chemistry model, the NMMB-MONARCHv1.0 model, to quantify the fraction of BrC from primary and secondary origin, and the contribution of this light-absorbing SOA on the radiative forcing of OA. We also propose to conduct a specific field campaign in the northeast Spain to study the relevance of this new chemistry in a region with high ammonia and ozone concentrations. Surface concentration measurements of ammonia, ozone and aerosol chemical composition will be conducted together with measurements of optical properties (absorption and scattering). Such dataset will provide for the first time unique information on the light-absorbing contribution of secondary BrC in northeast Spain.

KEY WORDS: Atmospheric chemistry, organic aerosols, ammonia, absorption, modeling

Parte B: INFORMACIÓN ESPECÍFICA DEL EQUIPO

B.1. FINANCIACIÓN PÚBLICA Y PRIVADA (PROYECTOS Y/O CONTRATOS DE I+D+I) DEL EQUIPO DE INVESTIGACIÓN (repita la secuencia tantas veces como se precise hasta un máximo de 10 proyectos y/o contratos).

1. Investigador del equipo de investigación que participa en el proyecto/contrato (nombre y apellidos): Enza Di Tomaso
   Referencia del proyecto: ERA4CS joint call 2016 (Topic B)
   Título: Dust Storms Assessment for the development of user-oriented Climate Services in Northern Africa, Middle East and Europe (DustClim)
   Investigador principal (nombre y apellidos): Dr. Sara Basart
   Entidad financiadora: European Research Area for Climate Services (ERA4CS)
   Duración: 01/09/2017-30/09/2020
   Financiación recibida (en euros): 319.125€
   Relación con el proyecto que se presenta: está muy relacionado
   Estado del proyecto o contrato: concedido

2. Investigador del equipo de investigación que participa en el proyecto/contrato (nombre y apellidos): Oriol Jorba, Enza Di Tomaso
   Referencia del proyecto: AXA Chair 2015
   Título: AXA Chair on Sand and Dust Storms
   Investigador principal (nombre y apellidos): Dr. Carlos Pérez García-Pando
   Entidad financiadora: AXA Research Fund
   Duración: 01/10/2016-30/08/2031
   Financiación recibida (en euros): 1.587.906,57€
   Relación con el proyecto que se presenta: está muy relacionado
   Estado del proyecto o contrato: concedido

3. Investigador del equipo de investigación que participa en el proyecto/contrato (nombre y apellidos): Oriol Jorba, Enza Di Tomaso, Francesco Benincasa
   Referencia del proyecto: 654169
   Título: Aerosols, Clouds, and Trace gases Research InfraStructure (ACTRIS-2)
   Investigador principal (nombre y apellidos): Dr. Gelsomina Pappalardo
   Entidad financiadora: H2020 European Comission
   Duración: 01/05/2015-30/04/2019
   Financiación recibida (en euros): 90.000€ (total: 9.541.215€)
   Relación con el proyecto que se presenta: está muy relacionado
   Estado del proyecto o contrato: concedido

4. Investigador del equipo de investigación que participa en el proyecto/contrato (nombre y apellidos): Oriol Jorba, Enza Di Tomaso
   Referencia del proyecto: CGL-2013-46736-R
   Título: Aerosol forecasting and assessment of radiative forcing on weather and climate applications with the online NMMB/BSC-CTM model
   Investigador principal (nombre y apellidos): Dr. Oriol Jorba
   Entidad financiadora: MINECO
   Duración: 01/01/2014 - 31/12/2016
   Financiación recibida (en euros): 170.610€
   Relación con el proyecto que se presenta: está muy relacionado
   Estado del proyecto o contrato: concedido

5. Investigador del equipo de investigación que participa en el proyecto/contrato (nombre y apellidos): Oriol Jorba
   Referencia del proyecto: 747048
   Título: Aerosol and Climate Response to NH3 in the NMMB/BSC Inter-Scale Model (ACRoNNIM)

Investigador principal (nombre y apellidos): Dr. Matthew Dawson
Entidad financiadora: H2020-MSCA-IF-2016
Duración: 01/07/2017 - 31/07/2019
Financiación recibida (en euros): 170.121,60€
Relación con el proyecto que se presenta: mismo tema
Estado del proyecto o contrato: concedido

6. Investigador del equipo de investigación que participa en el proyecto/contrato (nombre y apellidos): Oriol Jorba
Referencia del proyecto: CGL-2008-02818
Título: Implementación de un mecanismo químico acoplado “on-line” dentro del modelo atmosférico NMMB/DREAM global-regional
Investigador principal (nombre y apellidos): Dr. Oriol Jorba
Entidad financiadora: MICINN
Duración: 01/01/2009 - 12/31/2011
Financiación recibida (en euros): 127.292€
Relación con el proyecto que se presenta: está muy relacionado
Estado del proyecto o contrato: concedido

7. Investigador del equipo de investigación que participa en el proyecto/contrato (nombre y apellidos): Oriol Jorba, Enza Di Tomaso
Referencia del proyecto: SEV-2011-00067
Título: Severo-Ochoa
Investigador principal (nombre y apellidos): Dr. Mateo Valero
Entidad financiadora: Gobierno de España
Duración: 31/12/2011 - 31/12/2015
Financiación recibida (en euros): 4.000.000€
Relación con el proyecto que se presenta: está muy relacionado
Estado del proyecto o contrato: concedido

8. Investigador del equipo de investigación que participa en el proyecto/contrato (nombre y apellidos): Oriol Jorba
Referencia del proyecto: 157/PC08/3-12.0
Título: CALIOPE: Sistema de Calidad del aire operativo para España
Investigador principal (nombre y apellidos): Dr. Jose Baldasano
Entidad financiadora: Ministerio de Medio Ambiente
Duración: 01/01/2006 - 30/06/2010
Financiación recibida (en euros): 533.214€
Relación con el proyecto que se presenta: está muy relacionado
Estado del proyecto o contrato: concedido

9. Investigador del equipo de investigación que participa en el proyecto/contrato (nombre y apellidos): Francesco Benincasa, Oriol Jorba
Referencia del proyecto: N/A
Título: Convenio para la gestión y mantenimiento del Centro Regional para el Norte de África, oriente Próximo y Europa del Sistema de Evaluación y Aviso de Tormentas de Polvo y Arena de la OMM.
Investigador principal (nombre y apellidos): Dr. Francisco Doblas-Reyes
Entidad financiadora: AEMET (Agencia Estatal de Meteorología)
Duración: 02/05/2016-01/05/2018
Financiación recibida (en euros): 133.044€
Relación con el proyecto que se presenta: está algo relacionado
Estado del proyecto o contrato: concedido

10. Investigador del equipo de investigación que participa en el proyecto/contrato (nombre y apellidos): Oriol Jorba
Referencia del proyecto: 308395

Título: APPRAISAL - Air Pollution Policies foR Assesement of Integrated Strategies At regional and Local scales
Investigador principal (nombre y apellidos): Dr. Maria Luisa Volta
Entidad financiadora: Comisión europea. FP7-ENV-2012-one-stage
Duración (fecha inicio - fecha fin, en formato DD/MM/AAAA): 01/06/2012 - 31/05/2015
Financiación recibida (en euros): 39.891,64€ (total: 999.990€)
Relación con el proyecto que se presenta: está algo relacionado
Estado del proyecto o contrato: concedido

B.2. RELACIÓN DE LAS PERSONAS NO DOCTORES QUE COMPONEN EL EQUIPO DE TRABAJO (se recuerda que los datos de los doctores del equipo de trabajo y de los componentes del equipo de investigación no se solicitan aquí). Repita la siguiente secuencia tantas veces como precise.

No non-PhD researchers are included in the working group.

Parte C: DOCUMENTO CIENTÍFICO. Máximo 20 páginas.

C.1. PROPUESTA CIENTÍFICA

C.1.1. Antecedentes y estado actual

Brown Carbon as a significant absorbing aerosol

Atmospheric aerosols have a significant influence on the climate system. On average, aerosols cool the atmosphere by directly scattering solar radiation and indirectly through aerosol-cloud interactions (Myhre et al., 2013). However, some aerosol components are able of absorbing visible solar radiation and warming the lower atmosphere. The most well known type of absorbing aerosols are black carbon (BC; Haywood and Boucher, 2000) and mineral dust (Usher et al., 2003). Most organic aerosols (OA) can be characterized as "white" because they efficiently scatter visible radiation. Recently, optical and thermal analysis (e.g., Kirchstetter et al., 2004) and electron microscopy (e.g., Alexander et al., 2008) from laboratory and field experiments have provided strong evidence for the existence of some OA with light absorbing properties. In recent scientific literature, the term "Brown Carbon" has emerged to describe this type of OA, characterized by an absorption spectrum that smoothly increases from the vis to UV wavelengths (Kirchstetter et al., 2004; Laskin et al., 2015). All the aerosols capable of absorbing visible radiation tend to reduce the magnitude of the cooling forcing attributed to aerosols (Moosmüller et al. 2009). The climate response to aerosol absorption is found to depend strongly on the altitude of the absorbing aerosols (Hodnebrog et al., 2014; Ban-Weiss et al., 2012; Koch and Del Genio, 2010). In this sense, BrC constitutes a significant part of absorbing carbonaceous aerosols, especially at high altitudes (Zhang et al., 2017). Therefore, information on the spatial and vertical distribution of BrC is essential to improve the estimation of its climate impact.

Current climate models often assume that BC and mineral dust are the only two significant types of light-absorbing aerosols on the global scale, and they treat OA as a purely scattering component that leads to climate cooling (Bond et al., 2011; Ma et al., 2012). Feng et al. (2013) estimated for the first time the enhanced absorption of solar radiation due to BrC, only considering primary sources, in a global model. Their results suggest that the inclusion of BrC absorption produces a forcing of +0.11Wm−2 for a strongly absorbing BrC and of +0.04Wm−2 for a moderately absorbing BrC at the TOA. Such findings change the global radiative forcing of organic carbonaceous aerosols from cooling (−0.08Wm−2) to warming (+0.025Wm−2), with much larger impact on regional climate. Thus, climate simulations may introduce significant source of uncertainty if not considering the absorbing fraction of the organic aerosol.

Recent studies have shown that BrC is widespread in specific geographic areas and urban environments and makes a significant contribution to the total aerosol absorption (Kirchstetter and Thatcher 2012; Ramanathan et al. 2007). Such findings highlight the need for improved understanding of OA absorption properties, providing motivation for laboratory and field studies focused on fundamental understanding of the chemistry of BrC and its absorption properties (Laskin et al., 2015).

Secondary formation of Brown Carbon and aging effect of ammonia on SOA

Until recently, it was believed that BrC is produced solely by primary sources, which include residential coal combustion and biomass burning (Andreae and Gelencser 2006; Moosmüller et al. 2009). Recent studies have challenged this view and postulated the existence of various secondary sources of BrC resulting from multi-phase reactions between gas-phase, particulate and cloud micro-droplet constituents. Examples include the reactions of OH radicals with aromatic hydroxyacids and phenols in cloud water (Gelencser et al., 2003), aqueous reactions of glyoxal and methylglyoxal with ammonium sulfate (Galloway et al., 2009), gas-to-particle uptake of glyoxal by deliquesced ammonium-sulfate aerosol (Trainic et al., 2011), aqueous reactions between glyoxal and amino acids (De Haan et al., 2009), reactions of limonene SOA with ammonia (NH3) (Laskin et al., 2010) and with ammonium-sulfate (Bones et al., 2010), heterogeneous reactions of gaseous isoprene on acidic aerosol particles (Limbeck et al., 2003), and aqueous photochemistry of pyruvic acid in the presence of common atmospheric electrolytes (Rincon et al., 2009). Currently, the amount of additional

SOA mass and the climate effects of nitrogen-containing organic compounds produced by ammonia-driven secondary sources are highly uncertain, and require urgent attention from the atmospheric research community (Updyke et al., 2012).

The effect of added ammonia on the yields and important properties of SOA was previously investigated only in a limited number of chamber and field experiments. Na et al (2007) observed a significant increase in the SOA yield in the α-pinene+O3 SOA in the presence of ammonia. Huang et al (2012) also observed that ammonia significantly increased the yield of SOA from reactions between indoor ozone and VOCs emitted from cleaning products. However, for an aromatic SOA precursor styrene, the effect was opposite: the SOA yield decreased because NH3 induced efficient decomposition of the major SOA forming product, 3,5-diphenyl-1,2,4-trioxolane (Na et al. 2006). Lin et al. (2013) found no strong association between isoprene derived isoprene SOA and ammonia in samples collected in the southeastern US. Updyke et al. (2012) exposed a number of different types of SOA generated from biogenic and anthropogenic precursors to ppb levels of ammonia and found that light-absorbing nitrogen-containing organic compounds were produced inside SOA material. Clearly the effects of ammonia on SOA yields are complex and potentially dependent on the nature of the SOA precursor, clearly justifying a more systematic study of the ammonia-SOA aging chemistry.

Ammonia emissions are expected to increase worldwide in all future Earth scenarios because of the intensifying agricultural use of ammonia-based fertilizers (FAO, 2011). In addition, increases in temperature of 2-4 ºC due to climate change could lead to overall increases of up to 10-27% in ammonia emissions (Skjøth and Geels 2012). At the same time, the emissions of VOC precursors are also expected to increase because of the rising trends in the global temperatures (Constable et al., 1999). The combination of these two trends is likely to lead to significant changes in the mass-concentrations and chemical composition of SOA. Both experimental and modeling efforts are required to address this critical gap in our understanding of the effect of ammonia on SOA and resulting BrC radiative forcing.

Aim of the proposal

Under this scientific context, the present proposal aims to address the following scientific questions:

1) *What is the contribution of Brown Carbon and the novel chemistry on SOA aging with ammonia to the total radiative forcing of organic aerosols?*

2) *How does secondary formation of Brown Carbon contributes to total aerosol mass load and air quality?*

3) *What is the relevance of this novel chemistry on SOA aging in ambient field conditions?*

In order to address these scientific questions a multidisciplinary approach is planned combining expertise in aerosol modeling, laboratory experiments, field campaign retrievals, and climate assessment. The present proposal will allow the research team from the Atmospheric Composition group of Barcelona Supercomputing Center (AC-BSC), which has a wide experience in air quality and aerosol modeling from local to global scales, strengthen the established collaborations with: 1) the University of California, Irvine (UCI) research groups on aerosols photochemistry and modeling, 2) the Environmental Geochemistry Atmospheric Research IDAEA-CSIC group (EGAR), expert in field measurements of aerosol composition in Spain and Europe, and 3) the climate research group in NASA-Goddard Institute for Space Studies (NASA-GISS). Prof. Donald Dabdub and Prof. Sergey Nizkorodov (UCI), Dr. Marco Pandolfi (EGAR) and Kostas Tsigaridis (NASA-GISS) researchers will participate in the project as members of the working group. Their contribution will perfectly complement the modeling expertise of AC-BSC with lab-based research on SOA (UCI), ambient field measurements experience and analysis (EGAR), and climate assessments of atmospheric aerosols (NASA-GISS).

Background of the AC-BSC Research Group and relation with other research groups

The AC-BSC research group has strong expertise in air quality and aerosol modeling. Several scientific publications and PhD thesis have been conducted over Barcelona, Spain,

Europe and at global scale studying the air quality and atmospheric composition (e.g., Jorba et al., 2004; Pineda et al., 2004; Jorba et al., 2008; Jiménez-Guerrero et al., 2008; Pérez et al., 2011; Jorba et al., 2012; Spada et al., 2013; Badia and Jorba, 2015; Badia et al., 2017). The group coordinated a national initiative that aimed to develop an air-quality forecasting system for SPAIN under the umbrella of the CALIOPE project (Baldasano et al., 2008; Pay et al., 2010; Baldasano et al., 2011; Pay et al., 2012; Basart et al., 2012a). CALIOPE system is nowadays providing air quality forecasts for Europe at 12km, Spain at 4 km, and Andalucía, Cataluña and Madrid domains at 1km. Furthermore, works of the group (Pérez et al., 2006ab; Basart et al., 2009; Haustein et al., 2009; Basart et al., 2012b) have focused on the evaluation of mineral dust models with lidar observations, sun-photometer data and satellite imagery, and further improvements with the inclusion of dust radiative effect have shown a way to improve numerical weather prediction results through feedbacks between aerosol concentration and meteorology (Pérez et al., 2006a).

Since 2008, the group has been developing a new online chemical weather prediction system, the model NMMB-MONARCHv1.0 model (Pérez et al., 2011; Haustein et al., 2012; Jorba et al., 2012; Spada et al., 2013; Badia et al., 2017; DiTomasso et al., 2017), previously known as NMMB/BSC-CTM or NMMB/BSC-Dust. The model is a new fully on-line chemical weather prediction system for meso to global scale applications, and it is the main modeling framework of the present proposal. The NMMB-MONARCHv1.0 has been developed under the umbrella of several national research projects of the Ministry of Economy and Competiveness (CGL2006-11879/CLI, CGL2008-02818/CLI, CGL2010-19652 and CGL2013-46736-R1). The Principal Investigator (PI) of the present proposal has been the PI of two of these national projects. Nowadays, the NMMB-MONARCHv1.0 provides operational regional mineral dust forecasts to the World Meteorological Organization (WMO) Barcelona Dust Forecast Center (BDFC) and the Sand and Dust Storm Warning Advisory and Assessment System (SDS-WAS) Northern Africa-Middle East-Europe (NA-ME-E) Regional Center, both centers managed by a consortium between AEMET and BSC. Additionally, global aerosol forecasts with NMMB-MONARCHv1.0 are provided to the International Cooperative for Aerosol Prediction (ICAP) initiative (Sessions et al., 2016). Finally, the model will be the next generation air quality model for the CALIOPE system, replacing the current system based on WRF-CMAQ models.

The AC-BSC group has established several collaborations with national research teams: AEMET, CIEMAT, IDAEA-CSIC, CEAM, Technical University of Catalonia, University of Murcia. On an international level regular collaborations are established with: University of Aveiro, National Observatory of Athens, National Meteorological Agency of Turkey, National Centers for Environmental Predictions (NCEP; USA), NASA Goddard Institute for Space Studies (GISS, USA), International Research Institute for climate and Society (IRI, USA), University of California Irvine (UCI; USA). It is believed that such collaborations strongly benefit the progress of the scientific projects undertaken by AC-BSC related with the developments of the model NMMB-MONARCHv1.0. Furthermore, the group will coordinate the new European COST Action "International Network to Encourage the Use of Monitoring and Forecasting Dust Products" leaded by Dr. Sara Basart, member of the AC-BSC group.

Of particular relevance is the collaboration of AC-BSC with the University of California, Irvine. Strong synergies have been created with the Computational Environmental Sciences Lab managed by Prof. Donald Dabdub, expert in air quality and secondary organic aerosol modeling (Pay et al., 2010; Jorba et al., 2012; Gonçalves et al., 2012; Badia et al., 2017). Currently, Prof. Donald Dabdub and Prof. Sergey Nizkorodov (Department of Chemistry, UCI) are conducting laboratory and modeling experiments to characterize ammonia-related SOA chemistry and physics (i.e., Aiona et al., 2017; Montoya et al., 2017). Their findings will be the basis for European and global modeling experiments planned in the present proposal to further advance on the effect of BrC secondary formation on air quality and climate.

Furthermore, AC-BSC has a long history of collaboration with the EGAR IDAEA-CSIC (Pandolfi et al., 2014; Jorba et al., 2013; Pay et al., 2012). EGAR features complete laboratory facilities and state-of-the-art instrumentation for the measurement and characterization of atmospheric aerosols and trace gases. Additionally, they have many

years of experience performing field measurements related to aerosol and trace gas species in urban and rural environments. Thus, EGAR group offers the ideal synergistic complement to the AC-BSC, and a field campaign if planned in the present proposal with its support.

Previous project

The present proposal is the natural continuation of the project CGL2013-46736-R1 lead by the PI of the present proposal. In CGL2013-46736-R1, the radiative effects of aerosols were implemented in NMMB-MONARCHv1.0, the mineral dust component of the model was improved, and the aerosol-radiation impact on meteorology was studied. The knowledge acquired during that project on aerosol properties (Obiso et al., 2017) and radiative effects (Gkikas et al., 2017) contribute to identifying Brown Carbon as the next important aerosol component to be considered in air quality and climate models.

Bibliography

Aiona, P. A., et al., 2017. Aqueous reaction of 4-oxopentanal with ammonium sulfate forms brown carbon through a 2-methyl pyrrole intermediate. Environ. Sci. Technol. in review.

Alexander, D. T. L., Crozier, P. A., and Anderson, J. R.: Brown carbon spheres in East Asian outflow and their optical properties, Science, 321, 833–836, doi:10.1126/science.1155296, 2008.

Andreae, M. O. and Gelencser, A., 2006. Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols. Atmos. Chem. Phys., 6, 3131-3148.

Badia, A. and Jorba, O., 2015. Gas-phase evaluation of the online NMMB/BSC-CTM model over Europe for 2010 in the framework of the AQMEII-Phase2 project. Atmos. Environ., 115, 657-669.

Badia, A. et al., 2017. Description and evaluation of the Multiscale Online Nonhydrostatic AtmospheRe CHemistry model (NMMB-MONARCH) version 1.0: gas-phase chemistry at global scale. Geosci. Model Dev., 10, 609-638, doi:10.5194/gmd-10-609-2017.

Baldasano J.M, et al., 2008. CALIOPE: An operational air quality forecasting system for Spain - First annual evaluation and ongoing developments. Adv. Sci. Res., 2, 89-98.

Baldasano, J. M., et al., 2011. An annual assessment of air quality with the CALIOPE modeling system over Spain. Atmos. Environ., 409, 2163-2178.

Ban-Weiss, G. A. et al., 2012. Dependence of climate forcing and response on the altitude of black carbon aerosols. Clim. Dynam. 38, 897-911.

Basart, S., et al., 2009. Aerosol characterization in Northern Africa, Northeastern Atlantic, Mediterranean Basin and Middle east from direct-sun AERONET observations. Atmos. Chem. Phys., 9, 8265–8282.

Basart, S., et al., 2012a. Aerosols in the CALIOPE air quality modelling system: evaluation and analysis of PM levels, optical depths and chemical composition over Europe. Atmos. Chem. Phys, 12, 3363-3392.

Basart, S., et al., 2012b. Development and evaluation of the BSC-DREAM8b dust regional model over Northern Africa, the Mediterranean and the Middle East. Tellus B, 64, 1-23.

Bond, T. C. et al., 2011. Quantifying immediate radiative forcing by black carbon and organic matter with the Specific Forcing Pulse. Atmos. Chem. Phys., 11, 1505.

Bones, D.L. et al., 2010. Appearance of strong absorbers and fluorophores in limonene-O3 secondary organic aerosol due to NH4+-mediated chemical aging over long time scales. J. Geophys. Res. D 115 (D5), D05203. http://dx.doi.org/10.1029/2009JD012864.

Constable, J. V. H. et al., 1999. Modelling changes in VOC emission in response to climate change in the continental United States. Global Change Biology, 5, 791–806.

De Haan, D.O. et al., 2009. Secondary organic aerosol-forming reactions of glyoxal with amino acids. Environ. Sci. Technol. 43, 2818-2824.

Di Tomaso et al., 2017. Assimilation of MODIS Dark Target and Deep Blue observations in the dust aerosol component of NMMB-MONARCH version 1.0. Geosci. Model Dev., 10, 1107-1129, https://doi.org/10.5194/gmd-10-1107-2017.

FAO, 2011. Current world fertilizer trends and outlook to 2015. Food and Agriculture Organization of the United Nations. Rome, Italy, 2011.

Feng, Y. et al., 2013. Brown Carbon: a significant atmospheric absorber of solar radiation? Atmos. Chem. Phys., 13, 8607-8621.

Galloway, M.M. et al., 2009. Glyoxal uptake on ammonium sulphate seed aerosol: reaction products and reversibility of uptake under dark and irradiated conditions. Atmos. Chem. Phys., 9, 3331-3345.

Gelencser, A. et al., 2003. In-situ formation of light-absorbing organic matter in cloud water. J. Atmos. Chem., 45, 25-33.

Gkikas et al. Direct radiative effects of intense Mediterranean desert dust outbreaks. Atmos. Chem. Phys. (in preparation).

Gonçalves, M. et al., 2012. Impact of HONO sources on the performance of mesoscale air quality models. Atmos. Environ., 54, 168-176.

Haustein, K. et al., 2009. Regional dust model performance during SAMUM2006. Geophys. Res. Lett., 36.

Haustein, K., et al., 2012. Atmospheric dust modeling from meso to global scales with the online NMMB/BSC-Dust model – Part 2: Experimental campaigns in Northern Africa. Atmos. Chem. Phys., 12, 2933-2958.

Haywood, J., Boucher, O., 2000. Estimates of the direct and indirect radiative forcing due to tropospheric aerosols: a review. Rev. Geophys. 38 (4), 513-543.

Hodnebrog, Ø et al., 2014. How shorter black carbon lifetime alters its climate effect. Nat. Commun. 5, 5065.

Huang, Y. et al., 2012. Effect of ammonia on ozone-initiated formation of indoor secondary products with emissions from cleaning products. Atm. Environ., 59, 224-231.

Jiménez-Guerrero, P., et al., 2008. The Use of a Modelling System as a Tool for Air Quality Management: Annual High-Resolution Simulations and Evaluation. Sci. Tot. Environ., 390, 323-340.
Jorba, O. et al., 2004. Cluster analysis of 4-day back trajectories arriving in the Barcelona area, Spain, from 1997 to 2002. J. of Appl. Metorol., 43, 887-901.
Jorba, O., et al., 2008. Linking the advanced research WRF meteorological model with the CHIMERE chemistry-transport model. Environ. Model. & Soft., 23, 1092-1094.
Kirchstetter, T. W. and Thatcher, T. L., 2012. Contribution of organic carbon to wood smoke particulate matter absorption of solar radiation. Atm. Chem. Phys., 12, 6067-6072.
Jorba, O., et al., 2012. Potential significance of photoexcited NO2 on global air quality with the NMMB/BSC Chemical Transport Model. J. Geophys. Res., 117, D13301.
Jorba, O., et al., 2013. Overview of the meteorology and transport patterns during the DAURE field campaign and their impact to PM observations. Atmos. Environ., 77, 607–620.
Koch, D. and Del Genio, A. D., 2010. Black carbon semi-direct effects on cloud cover: review and synthesis. Atmos. Chem. Phys. 10, 7685-7696.
Laskin, A., Laskin, J., and S. A., Nizkorodov, 2015. Chemistry of Atmospheric Brown Carbon. Chem. Rev., 115, 4335-4382, doi:10.1021/cr5006167.
Laskin, J. et al., 2010. High-resolution desorption electrospray ionization mass spectrometry for chemical characterization of organic aerosols. Analyt. Chem., 82, 2048-2058.
Limbeck, A., Kulmala, M., Puxbaum, H., 2003. Secondary organic aerosol formation in the atmosphere via heterogeneous reaction of gaseous isoprene on acidic particles. Geophys. Res. Lett. 30 (19), 1996. http://dx.doi.org/10.1029/2003GL017738.
Lin, Y. H. et al., 2013. Investigating the influences of SO2 and NH3 levels on isoprene-derived secondary organic aerosol formation using conditional sampling approaches. Atmos. Chem. Phys., 13, 8457-8470.
Ma, X. et al., 2012. Aerosol direct radiative forcing based on GEOS-Chem-APM and uncertainties. Atmos. Chem. Phys., 12, 5563.
Montoya, J. et al., 2017. Secondary Organic Aerosol from Atmospheric Photooxidation of Indole. Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-270, in review.
Moosmüller, H. et al., 2009. Aerosol light absorption and its measurement: A review. J. Quant. Spect. and Rad. Tr., 110, 844-878.
Myhre, G. et al. in Climate Change 2013: The Physical Science Basis (eds Stocker, T. F. et al.) 659-740 (IPCC, Cambridge Univ. Press, 2013).
Na, K., Song, C. and Cocker, D. R., 2006. Formation of secondary organic aerosol from the reaction of styrene with ozone in the presence and absence of ammonia and water. Atmos. Environ., 40, 1889-1900.
Na, K. et al., 2007. Effect of Ammonia on Secondary Organic Aerosol Formation from α-Pinene Ozonolysis in Dry and Humid Conditions. Environ. Sci. & Tech., 41, 6096-6102.
Obiso, V. et al., 2017. Impact of aerosol physical properties on mass scattering cross sections. J. Aerosol Science. Accepted.
Pandolfi, M. et al., 2014. Effects of sources and meteorology on particulate matter in the Western Mediterranean Basin: An overview of the DAURE campaign. J. Geophys. Res., 119, 4978 - 5010.
Pay, M.T., et al., 2010. A full year evaluation of the CALIOPE-EU air quality modeling system over Europe for 2004. Atmos. Environ., 44 (27): 3322-3342.
Pay, M. T., et al., 2012. Assessing sensitivity regimes of secondary inorganic aerosol formation in Europe with the CALIOPE-EU modeling system. Atmos. Environ., 46, 376–396.
Pérez, C., et al., 2006a. Interactive dust-radiation modelling: a step to improve weather forecast. J. Geophys. Res., 111, D16206.
Pérez, C., et al., 2006b. A long Saharan dust event over the western Mediterranean: Lidar, sun photometer observations and regional dust modelling. J. Geophys. Res., 111, D15214.
Pérez, C., et al., 2011. Atmos. Chem. Phys., 11, 13001-13027.
Pineda, N. et al., 2004. Using NOAA AVHRR and SPOT VGT data to estimate surface parameters: application to a mesoscale meteorological model. Int. J. of Remote Sensing, 25, 129-143.
Ramanathan, V. et al., 2007. Warming trends in Asia amplified by brown carbon cloud solar absorption. Nature, 448, 575-578.
Rincon, A.G. et al., 2009. Optical absorptivity versus molecular composition of model organic aerosol matter. J. Phys. Chem. A, 113, 10512-10520.
Sessions, W. R. et al., 2015 Development towards a global operational aerosol consensus: basic climatological characteristics of the International Cooperative for Aerosol Prediction Multi-Model Ensemble (ICAP-MME), Atmos. Chem. Phys., 15, 335-362, https://doi.org/10.5194/acp-15-335-2015.
Skjøth C.A., Geels C., 2013. The effect of climate and climate change on ammonia emissions in Europe. Atmos. Chem. Phys., 13, 117–128.
Spada, M. et al., 2013. Modeling and evaluation of the global sea-salt aerosol distribution: sensitivity to size-resolved and sea-surface temperature dependent emission schemes. Atmos. Chem. Phys., 13, 11735-11755, https://doi.org/10.5194/acp-13-11735-2013.
Trainic, M. et al., 2011. The optical, physical and chemical properties of the products of glyoxal uptake on ammonium sulfate seed aerosols. Atmos. Chem. Phys., 11, 9697-9707.
Updyke et al., 2012. Formation of brown carbon via reactions of ammonia with secondary organic aerosols from biogenic and anthropogenic precursors. Atm. Environ., 63, 22-31.
Usher, C.R., Michel, A.E., Grassian, V.H., 2003. Reactions on mineral dust. Chem. Rev.
103 (12), 4883-4939.
Zhang, Y. et al., 2017. Top-of-atmosphere radiative forcing affected by brown carbon in the upper troposphere. Nat. Geosciences.

C.1.2. Hipóstesis de partida, objetivos generales, adecuación

Initial hypothesis:

Organic aerosols (OA) affect climate forcing and human health, but still large uncertainties exist in their sources and evolution. OA makes up a large fraction of the submicron particulate matter, 20 to 90%. Recent studies have shown that secondary organic aerosols (SOA) accounts for a large fraction of the OA burden. Most OA efficiently scatter visible radiation and contribute to a cooling effect in the atmosphere. However, a significant fraction of OA absorbs radiation in the near-UV and visible ranges. Such fraction is known as "Brown Carbon" (BrC). BrC contributes together with Black Carbon (BC) and mineral dust to warming the atmosphere.

The initial hypotheses supporting the objectives of the project are:

1) Current climate models assume that BC and mineral dust are the only two significant types of light-absorbing aerosols on the global scale, and they treat OA as a purely scattering component leading to climate cooling. However, recent advances on the identification and characterization of BrC indicate that BrC may contribute substantially the total aerosol absorption at specific wavelengths. The inclusion of BrC in atmospheric composition models may provide new insights on the warming effect of OA at global scale.
2) BrC is directly emitted during wood and fossil fuel combustion and it can also be formed by atmospheric oxidation of gas-phase species as a secondary pollutant. The light-absorbing properties in some SOA are observed after aging processes in the formed aerosol. The browning of SOA may be a significant contributor to the total absorption of BrC.
3) Recent studies have identified that some initially scattering SOA undergoes browning during exposure to reduced nitrogen compounds such as ammonia, ammonium from dissolved ammonium sulfate or other salts, and amino acids. The contribution of such light-absorbing OA to the total warming effect of BrC is still highly uncertain and it may be significant in areas that have high emissions of ammonia or dissolved ammonium salts.
4) Several field campaigns have focused on the characterization of OA. The DAURE campaign is a good example of such campaigns. It was conducted in the northeast Spain, and its main objective was the study of the origin of the pollution episodes and the emissions, formation, transport, and transformation of aerosols, particular attention was devoted to quantitatively understand the sources and formation mechanisms of secondary inorganic and organic aerosols (SIA and SOA). Important knowledge was achieved during DAURE on OA processes. The results of DAURE represent an excellent framework to further investigate the role of SOA aerosols in the northeast Spain, but the measurements were conducted in locations characterized by low ammonia concentrations. In this sense, specific field measurements of optical properties (absorbing AOD, Single Scattering Albedo, absorbing Angstrom Exponent) in areas with high concentrations of ammonia and OA may contribute to a better understanding of the role of such browning processes of SOA in ambient conditions.

Main objective:

The main objective of this proposal is to explore the effect of the reactive uptake of ammonia (NH3) by secondary organic aerosols (SOA) on the optical properties and burden of anthropogenic and biogenic SOA using both experimental and modeling approaches. We propose implementing new NH3-SOA chemistry in a state-of-the-art atmospheric-chemistry model to quantify the fraction of BrC from primary and secondary origin, and the contribution of this light-absorbing SOA and BrC on the radiative forcing of OA. We also propose specific field campaign in the northeast Spain to study the relevance of this new chemistry in a region with high NH3 and O3 concentrations. Surface concentration measurements of NH3, O3 and OA will be conducted together with optical properties measurements of absorbing AOD, Single Scattering Albedo and absorbing Angstrom Exponent. Such dataset will provide unique information on the light-absorbing contribution of secondary BrC in northeast Spain.

Appropriateness to the National and European research strategy

The topic of the present proposal covers the *fifth societal challenges identified by the Spanish research strategy about Climate Change*. A better understanding about the role of light-absorbing organic aerosols on the radiative balance of atmospheric models will contribute to reduce the uncertainties on the quantification of the radiative forcings attributed

to anthropogenic and natural aerosols. Such advances will contribute in the definition of future mitigation and adaptation measures to face the challenges that Climate Change is creating. The *goals defined in the HORIZON 2020 on climate change and air quality modeling are in agreement with the strategy defined by the Spanish Government*. In this sense, the improvement and evaluation of air quality modeling system and the advancement in the anthropogenic forcings that are enhancing the climate change are key areas of research at the European level.

Additionally, the *objectives of the present proposal are in agreement with the research lines defined by the "Estrategia Española de Ciencia Tecnología e Innovación 2013-2020"* of the Ministry of Economy and Competitiveness. The Strategy identifies the aerosols as one of the critical pollutants to monitor and over which further modeling activities are required. One of the targets of the Plan is the emission reduction of precursors of secondary aerosols. Mostly 40% of PM10 is composed of secondary aerosols. In this sense, the modeling tools under development in the present project will provide valuable information about the chain production of secondary aerosols and will allow identifying those mechanisms that may impact on their reduction in the atmosphere. *Concerning the actions to promote the research, the Strategy identifies air quality modeling as a key topic where the Spanish research community has to further increase its expertise*. Models need to be improved and evaluated under different conditions. The present proposal has as first objective to further understand the contribution of secondary organic aerosols to the total aerosol concentration and the impact of such aerosols on the radiative forcings affecting Climate.

Finally, the *Horizon 2020 2016-2017 Work Programme for the "Climate action, environment, resource efficiency and raw materials" Challenge* describes the European air quality economic sector as 'critical to moving forward the transition to a circular economy, and also an important source of growth and jobs.' Additionally, *climate action is one of the primary crosscutting actions highlighted by the Horizon 2020 program, and the effect of aerosol species on Earth's radiative balance represents the largest uncertainty in predicting the trajectory of climate change*. Thus, the expected impact of this research, to improve the quality of predictions of aerosol mass and optical properties, and their impact on air quality and climate in one of the premier Spanish chemical weather prediction systems, NMMB-MONARCHv1.0, falls perfectly in line with European research priorities and concurrent initiatives.

C.1.3. Objetivos específicos

To achieve the main objective of the project *three specific objectives are defined*:

1) Explore the *role of recently identified chemistry of NH3-related SOA formation and aging on the total mass of BrC, OA and particulate matter* by means of atmospheric-chemistry modeling experiments conducted with the model NMMB-MONARCHv1.0.

a. Implementation in the atmospheric-chemistry model a new organic aerosol tracer tagged as "Brown Carbon". Inclusion of emission, transport and deposition of the new aerosol component.

b. Incorporate new NH3-related SOA formation and aging chemistry proposed by collaborators at the University of California, Irvine, in the aerosol chemical mechanism of the NMMB-MONARCHv1.0 model.

c. Evaluation of the new chemistry of BrC with regional modeling experiments over the northeast Spain at high-horizontal and temporal resolution (1 km^2 and hourly frequency). The period of study will cover July 2009 to study the dataset from the DAURE summer campaign, and July 2019 to study the specific dataset obtained from the field campaign proposed in the present project. Additionally, a global modeling experiment with the new chemistry will contribute to the AEROCOM-Phase III initiative.

2) Conduct a field campaign in the northeast Spain to characterize the surface concentrations of oxidants and reactants associated to light-absorbing aging of SOA during July 2019. The technical measurements will be sub-contracted to the Environmental Geochemistry and Atmospheric Research (EGAR) group of IDAEA-CSIC. Measurements of

NH3, O3, OA and aerosol optical properties will be retrieved in Vic, an area downwind Barcelona urban area characterized by high O3 and NH3 concentrations and surrounded by forest and countryside landscape.

a. Design of the field campaign based on previous results of the DAURE experimental campaign conducted by the EGAR group. Selection of the specific location and instruments to be deployed.

b. Conduct the field campaign. Support to the EGAR group on the technical tasks of the measurements.

c. Analysis of the results and characterization of the absorption and OA concentrations measured. Identification of related NH3-SOA chemistry in Vic compared with data from Montseny and Barcelona.

3) Quantify the contribution of BrC to the radiative forcing of Organic Aerosols by means of modeling experiments.

a. Implement the radiative effect of BrC aerosol on the radiation parameterization of the NMMB-MONARCHv1.0 to explicitly solve the radiative forcing of such aerosol component.

b. Characterization of the optical properties of BrC (extinction efficiency, single scattering albedo and asymmetry parameter) by means of a Mie or T-Matrix code.

c. Model experiment to quantify the radiative forcing of BrC at global and regional scale.

C.1.4. Metodología

For the present project 4 persons from the research team and 4 persons from the working team are envisaged to achieve the objectives proposed. Three external scientists experts in secondary organic aerosols will be part of the working team: Prof. Donald Dabdub (UCI), Prof. Sergey Nizkorodov (UCI), and Dr. Kostas Tsigaridis (NASA-GISS). Additionally, another external scientist expert in optical properties of aerosols will be also part of the working team, Dr. Marco Pandolfi (EGAR). Complementing the team, 1 Post-doc and 1 Technician will be hired. The Post-doc1 will have experience on secondary aerosol modeling and will contribute to WP1 and WP2 implementing and assessing the impact of new BrC chemistry. The Technician will be an IT expert on parallel programming and will contribute to WP1 and WP3 in the coding and optimization of new parallel routines regarding the new chemistry and optical properties.

In order to achieve the proposed objectives, we define the following methodology and working plan:

Work package 1: BrC chemistry and role of the reactive uptake of NH3 by SOA: model development and simulations

The WP1 will cover the specific objective 1. The BrC chemistry will be implemented in the NMMB-MONARCHv1.0 model. Collaborators from University of California, Irvine, Prof. Donald Dabdub and Prof. Sergey Nizkorodov will contribute on the design of the related NH3-SOA chemistry to be included in the model.

Task 1.1 Review of BrC chemistry and secondary organic aerosol aging

The proper treatment of BrC in chemical transport models requires a deep overview on the different formation mechanisms. Conventional knowledge associates light-absorbing OA with primary sources and light-scattering OA with secondary sources. Primary BrC sources well known are smoldering forest fires and residential coal combustion. However, recent works have demonstrated the existence of secondary sources of BrC via multi-phase reactions between gas-phase, particulate-phase and cloud micro-droplet constituents. Furthermore, browning transformation has been observed in specific SOA ageing processes. This task will further identify the most relevant sources and chemistry pathways of BrC to be implemented in NMMB-MONACHv1.0 model in order to be ready to study the role of novel aging chemistry of BrC-SOA.

Task 1.2 Implementation of BrC aerosol component in NMMB-MONARCHv1.0 model

The NMMB-MONARCHv1.0 has an aerosol mechanism that models the life cycle of the most relevant aerosol components in the troposphere: mineral dust, sea salt, organic aerosol, black carbon, sulfate, nitrate and ammonium. In this approach, the organic aerosol (OA) represents all type of primary and secondary organic carbon with no distinction on their optical properties. Aerosol models are constrained by optical measurements, mainly Aerosol Optical Depth (AOD) and abosrbing AOD. To calculate the total AOD from the mass concentration solved by NMMB-MONARCHv1.0, we assume some optical properties derived from OPAC. Concerning OA, OPAC provides the refractive index of a purely scattering OA. As discussed in Section C1, a significant fraction of the OA absorbs in the UV-vis range, and this have a strong impact on the radiative forcing of OA. The inclusion of an absorbing OA will strongly improve the effect of OA in the atmosphere, considering both its scattering and absorbing properties. In this sense, this task will add a new aerosol tracer in NMMB-MONACHv1.0, the Brown Carbon (BrC). The microphysical characteristics of BrC will be similar to OA, but a different refractive index will be assumed in order to consider the absorbing properties of BrC. All transport, sedimentation, and dry and wet deposition processes will be treated similar for both organic components.

The inclusion of an explicit BrC requires a new treatment of the OA emissions and chemistry. With the new scheme, the emissions of primary OA will be split in OA (purely scattering) and BrC (scattering and absorbing). The main primary emissions of BrC are from fossil fuel and biomass burning combustion, currently the particulate matter from such emissions are split in BC and OA. Now, the OA fraction will be split in 60-70% OA and 40-30% BrC, selecting a recommended split fraction from the literature.

Finally, the secondary formation of organic matter will also be split in BrC-SOA and default SOA. There are well known reactions that form purely scattering SOA, and others that form BrC-SOA. The most relevant formation pathways in the troposphere will be implemented for both SOA and BrC-SOA. The aging process that enhance light-absorption of SOA will be addressed in Task 1.3.

The expertise of the AC-BSC research team on numerical modeling makes this task completely affordable.

Task 1.3 Implementation of new NH3-SOA aerosol chemistry in NMMB-MONARCHv1.0 model

Prof. Sergey Nizkorodov and Prof. Donald Dabdub are currently performing chamber experiments designed to expand on their work to quantify the impact of NH3 on SOA aging using a variety of biogenic and anthropogenic SOA precursors under various atmospherically relevant temperature and relative humidity conditions. The work proposed in this task will collaboratively parameterize their published and on-going work on the formation of light-absorbing SOA from NH3 processing for inclusion into the NMMB-MONARCHv1.0 model. Current version of the model includes a simplified treatment of SOA with a two-product scheme that contributes to the total OA (purely scattering) mass. The new chemistry will expand the current implementation with a state-of-the-art BrC chemistry mechanism to quantify the role of the new NH3-SOA chemistry on the burden of OA. Considering that this new chemistry acts as a sink of NH3 not accounted in previous model simulations, it is expected that the burden of secondary inorganic aerosols will change (as suggested by preliminary results of Prof. Dabdub and Nizkorodov). Results in this direction will be discussed.

Prof. Sergey Nizkorodov and Prof. Donald Dabdub are already publishing their work regarding this new chemistry. This reduces the uncertainty in the feasibility of the task due to the lack of applicable results. The expertise of the AC-BSC research team on numerical modeling and the participation of Prof. Sergey Nizkorodov and Prof. Donald Dabdub makes this task completely affordable.

Task 1.4 Modeling experiments

Several numerical simulations with the new aerosol scheme implemented in NMMB-MONARCHv1.0 are planned. Two domains of study and periods of analysis are proposed:

a) Regional scale: High-resolution simulations will be conducted to explore the role of BrC chemistry in the northeastern Spain at a resolution of 1 km x 1 km. Anthropogenic emissions will be provided by the HERMESv3 emission inventory developed by BSC at hourly temporal resolution. Biomass burning emissions will be obtained from the CAMS Global Fire Assimilation System GFASv1.2. Biogenic emissions will be calculated online within NMMB-MONARCHv1.0 with the MEGANv2.04 model. Ground-based observations gathered in WP2 will be used to evaluate model performance on surface concentration of organic aerosols and constrain the total AOD and the absorbing AOD. Retrievals of Single Scattering Albedo and Absorbing Angstrom Exponent will be used to identify periods of time with presence of BrC. Two periods of study will be the focus of the research: July 2009 (summer DAURE campaign with no effect of NH3) and July 2019 (field campaign to be conducted in WP2 under strong NH3 conditions).

b) Global scale: The updated model will be configured for an evaluation of the impact of NH3 on SOA globally, at a resolution of 0.7º x 0.5º. Model input conditions, including gas and aerosol species emissions will be compiled from existing inventories: HTAPv2 for anthropogenic emissions, GFASv1.2 for biomass burning emissions, and MEGANv2.04 for biogenic emissions. Ground-based and satellite observations of key species and bulk aerosol optical properties will be used to evaluate model performance. Model results will be used to assess the impact of the newly incorporated chemistry on air quality hot spots and on the global distribution of absorbing AOD. An annual simulation of 2015 will be conducted and will contribute to the AEROCOM-Phase III global aerosol model intercomparison initiative.

The expertise of the AC-BSC research team in using supercomputer resources assures the success of the numerical experiments planned in this task.

Work package 2: Experimental campaign

The WP2 will cover the specific objective 2. An oriented field campaign is planned to identify the role of the NH3-SOA chemistry in the northeast Spain. Surface concentration of oxidants, reactants, OA and aerosol optical properties will be measured during a summer month. Considering the complexity of this objective, the technical measurements will be sub-contracted to the Environmental Geochemistry and Atmospheric Research (EGAR) group of IDAEA-CSIC, EPO of the proposal. EGAR group has a wide experience conducting measurement campaigns oriented to study aerosol sources, chemistry, and optical properties. Prof. Donald Dabdub and Prof. Sergey Nizkorodov will contribute on the design of the field campaign together with researchers of EGAR group.

Task 2.1 Review previous experimental campaigns on organics in Spain

A detailed revision of previous field campaigns conducted in Spain will be performed. The EGAR group has been measuring atmospheric pollution in the region at urban and rural sites with particular attention to aerosol composition. The group has a large database of measurements starting in 2000 and being updated since nowadays. From their database, two sets of data are of special interest for the present proposal: DAURE summer campaign, and series of simultaneous measurements of PM levels, chemical composition and optical properties of aerosols.

a) The DAURE campaign (Determination of the Sources of Atmospheric Aerosols in Urban and Rural Environments in the Western Mediterranean) was a multidisciplinary international field campaign aimed at investigating the sources and meteorological controls of PM in the Western Mediterranean Basin (WMB). Particular attention was devoted to quantitatively understand the sources and formation mechanisms of secondary inorganic and organic aerosols (SIA and SOA) and the effects of anthropogenic activities on SOA formation at local and regional levels. Several state-of-the-art methods such as ^{14}C analysis, proton-transfer reaction mass spectrometry (PTRMS) for volatile organic compounds (VOCs) and high-resolution time-of-flight aerosol mass spectrometry (HR-AMS) for fine-particle composition were applied for the first time in the WMB region as part of DAURE, together with more traditional filter and impactor-based methods.

b) Simultaneous measurements of PM levels, chemical composition and optical properties of aerosols. The EGAR group collected measurements of aerosol PM10 mass concentrations and optical properties (aerosol scattering and absorption properties with the combination of a Nephelometer, an Aethalometer and a Multi Angle Absorption Photometer) at a rural sites in NE of Spain (Montseny and Montsec sites). From this set of measurements the EGAR group derived intensive properties for the PM10 particles (scattering Angstrom exponent, absorption Angstrom exponent, asymmetry parameter, single scattering albedo, and mass scattering cross sections).

The two sets of measurements are of extreme value to study the BrC chemistry planned in this proposal. The main limitation identified from the initial analysis of the data is the lack of ammonia measurements. And, from EGAR group experience, the sites where they measure (Montseny and Montsec rural sites) low concentrations of ammonia are present. This is the main reason to propose a complementing field campaign in the present project. The support from Dr. Marco Pandolfi (EGAR) on the review of previous campaigns assure the creation of an excellent database to constrain our model.

Task 2.2 Design of the experimental campaign: measure of organic aerosol mass, NH3 concentration and aerosol optical properties

The browning transformation in some SOA (e.g., limonene/O3 SOA) has been observed during exposure to high-concentration of reduced nitrogen compounds (NH3, NH4+, and amino acids). This chemistry may contribute to aerosol absorption in ambient conditions where high concentration of reduced nitrogen compounds, frequent changes in relative humidity, and high photochemical activity are observed. Such conditions may exist in predominantly agricultural areas and in forested areas. In this sense, the selection of a location to conduct a field campaign with the aim to identify this chemistry should have particular characteristics: high oxidant concentrations (e.g., O3, OH), high NH3 concentrations, and to be in an agricultural area with high photochemistry activity.

In the NE of Spain, there is a place characterized by recurrent exceedances of ozone concentrations (above 200 μg m-3 of hourly concentration) during summer time and high concentrations of ammonia have been measured (40 μg m-3 of daily mean concentrations). It is located in a rural area where farming and agriculture activities are dominant. This site, the Plana de Vic, will be the location to conduct the field campaign. More specific selection of the place where to install the measurement instruments will be done during the first year of the project, when the field campaign will be designed in detailed with the collaboration of EGAR group; Dr. Marco Pandolfi (expert on optical properties) from EGAR, Prof. Sergey Nizkorodov (expert on BrC chemistry) and Prof. Donald Dabdub (expert on secondary organic aerosols modeling) from UCI will contribute in this process. The period for the measurements has to be during summer, where high concentrations of O3 and NH3 are observed.

Considering the long expertise of EGAR group on field measurements in the area of interest, the field campaign will be sub-contracted to them. The planned set of measurements and instruments to be retrieved and used have been discussed with Dr. Marco Pandolfi (EGAR) and EGAR group. The selected measurements are:

- *Standard monitors* of NH3, O3, NO2 and VOCs concentrations.
- *Filters* to analyze off-line the concentration of organic carbon, elemental carbon, sulfate, nitrate, ammonium, mineral fraction, trace elements like levoglucosan (tracer of organic mass from biomass burning origin), pH. It is planned to use a filter every 12h or 24h.
- *Multi-Angle Absorption Photometer* (MAAP, model 5012,Thermo Scientific) to measure loading of black carbon (BC) in the atmosphere. The aerosol light absorption coefficient (σap) at 637 nm will be obtained from BC mass concentrations assuming a constant mass absorption cross section (MAC) of 6.6m2 g-1 provided by the manufacturer.
- *Aethalometer instrument* (model AE-33 Magee Scientific) to measure aerosol light absorption coefficients (σap) at seven different wavelengths from UV to near IR (370, 470, 520, 590, 660, 880 and 950 nm). From the multi-wavelengths aethalometer measurements it will be possible to obtain the Absorption Angstrom Exponent (AAE) which is closely related to

aerosol composition and type, BrC aerosols yield AAE is over 2 while BC near 1. Thus, AAE can provide an indication of the dominant absorbing aerosol type such as dust, black carbon, brown carbon, or mixtures of them.
- *LED-based integrated Nephelometer* (model Aurora 3000, ECOTECH Pty, Ltd, Knoxfield, Australia) to measure aerosols light scattering (σsp) and hemispheric backscattering (σbsp) coefficients measured at three different wavelengths (450, 525 and 635 nm). From the multi-wavelengths nephelometer measurements it will be possible to obtain the Scattering Angstrom Exponent (SAE) which is closely related to the size of the sampled aerosols. From multi-wavelengths scattering and absorption measurements it will be possible to estimate the Single Scattering Albedo (SSA) from UV to the visible range.
- *Single-channel CHM15K (CHM) elastic lidar* (ceilometer) to measure the vertical structure of atmospheric aerosols through the profiles of the lidar signals. The instrument is designed for continuous cloud base height determination and aerosol backscatter coefficient measurements during the day and night. With the Ceilometers it is possible to obtain maps of the aerosol load in the atmosphere with high temporal and vertical resolution.
- *Aerosol Chemical Speciation Monitor* (ACSM) to provide quantitative measurements of the non-refractory components of submicron atmospheric aerosol: sulfate, nitrate, chloride, ammonium and organic aerosol (OA) with a time resolution of 15 min to 1 h. The instrument provides organic mass spectra, which can be analyzed by multivariate mathematical techniques, such as positive matrix factorization (PMF), to identify the OA origin or OA. The most common OA sources/types are: hydrocarbon-like OA (HOA), related to primary engine emissions; biomass burning OA (BBOA), related to primary emissions from biomass combustion; and oxygenated OA (OOA), attributed to secondary OA (SOA).

After the campaign, filters cutouts will be sent to Prof. Sergey Nizkorodov (UCI) for a further UV-vis absorption spectra measurement in his laboratory with a dual-beam spectrometer (Shimadzu UV-2450). The extent of SOA browning is quantified in terms of effective mass absorption coefficient (MAC) of the organic material. Wavelength-dependent MAC can be directly calculated from the base-10 absorbance of an SOA extract.

The feasibility of the campaign will strongly depend on the final funding or not of its cost. If the campaign is not funded, the AC-BSC group will focus the work in the DAURE campaign data and historical measurements of EGAR group.

Task 2.3 Summer campaign
The measurements will be conducted during July 2019. The research team of the proposal will support the EGAR group in the technical work required to perform the measurements. During the campaign, Prof. Sergey Nizkorodov and Prof. Donald Dabdub will be invited to Spain to participate in the measurements. A key element for a successful campaign is the weather. The research group will analyze the seasonal forecasts and mid-term weather forecasts to identify which weeks of July will be the best one to have high temperatures and high insolation in the area. Additionally, the CALIOPE forecasting system will be consulted during the campaign to forecast when the polluted air masses from Barcelona will affect the area and when high O3 concentrations may be expected.

To assure a proper success in the summer campaign, the technical work will be sub-contracted to EGAR group, with a wide experience in field campaigns, EPO of the proposal.

Task 2.4 Analysis of the data from the campaign
The results of the field campaign will be analyzed and used for model constraints the rest of the project. All the collaborators in the work team will be involved in the analysis and discussion of results together with EGAR group. A publication with the overview of the campaign will be prepared at the end of the project, and the data retrieved will be made available to the research community once the quality control and quality assurance of the data will be conducted.

Work package 3: Radiative forcing of organic aerosols
The WP3 will cover the specific objective 3. This work package aims at quantifying the contribution of BrC and secondary BrC to the radiative forcing of organic aerosols globally.

For that, the optical properties of OA and BrC will be revised and updated, and the effect of BrC in the radiative parameterization of NMMB-MONARCHv1.0 will be included. Dr. Kostas Tsigaridis from NASA-GISS will be deeply involved in this work package in the definition of the optical properties of BrC, the set-up of the model experiments, and the assessment of the radiative forcing of both OA and BrC.

Task 3.1 Review of aerosol optical properties: fresh, aged, coating and water uptake
Currently, the NMMB-MONARCHv1.0 model uses the OPAC database to quantify the optical properties of each aerosol component. This information is then used to compute the aerosol optical depth or the radiative forcing of aerosols. The refractive indexes from OPAC are outdated for some particular aerosols, i.e., mineral dust or organic aerosols, and water uptake effect are in underestimated, i.e., sulfates. In this task, we will review the optical properties for carbonaceous aerosols and their physico-chemical state (freshly emitted, aged, with coating of other aerosols such sulfate, dry or with water uptake).

Regarding organic aerosols, there are recent works quantifying their refractive index. To have an idea, the OA refractive index used in NMMB-MONARCHv1.0 is 1.53 + 0.008i at 300 nm and 1.53 + 0.006i at 550 nm. Such values are representative of mainly scattering organic aerosols with slight absorption properties. Some recent works quantifies the imaginary part of the refractive index for BrC ranging from <0.0001 to 0.2 at 550 nm. This large variability indicates that BrC in regions not dominated by BC may be a significantly contributor to the total absorption. Depending on the treatment of BrC on models, if any, may significantly change the radiative forcing of OA.

On the other hand, Black carbon (BC) is emitted from fossil and biofuel combustion, and biomass burning. Once in the atmosphere, the aerosol can be coated with soluble material. The optical properties of the aerosol can strongly vary from freshly emitted BC and aerosol with internal mixtures of black carbon, sulfate, organics and water. Depending on the treatment of BC internal mixtures, the mass absorption cross section may be enhanced by a factor of 1.8 to 2, from laboratory measurements, and a lower factor of 1.2 to 1.6 near source regions, from ambient BC. Similar to OA, but in a order of magnitude major, the treatment of BC as purely fresh or aged coated aerosol may have a strong impact on the absorption simulated by models.

A revision of the recent works quantifying the optical properties of aerosols will be conducted and a specific parameterization of the optical properties will be outlined to be implemented in NMMB-MONARCHv1.0 in task 3.2.

Task 3.2 Implementation of BrC effects on radiative parameterization of NMMB-MONARCHv1.0 model and update of aerosol optical properties
Aerosol-radiation interaction process was implemented in NMMB-MONARCHv1.0 (previously known as NMMB/BSC-CTM) from projects CGL2006-11879 (for dust) and CGL2013-46736 (for other relevant global aerosols sea-salt, black carbon, organic carbon and sulfate). Following the same approach, the effects of BrC will be implemented in this project. Additionally, the contribution of SOA to the radiative direct effect will be also implemented for non-absorbing and absorbing species. The NMMB-MONARCHv1.0 atmospheric driver uses the RRTMG radiative transfer model, which allows the inclusion of aerosol effects in the radiative budget computation. For each aerosol size bin and wavelength we will calculate the extinction efficiency, single-scattering albedo and asymmetry factor with a Mie or T-matrix code. Complex refractive indexes for aerosol components will be updated following Task 3.1. A weighted integration across the spectral bandwidth is done with the extraterrestrial solar irradiance spectrum for the solar wavelengths and the Planck function for long-wave wavelengths. Mean values of optical thickness, single-scattering albedo, and asymmetry factor are in turn derived for each spectral band and atmospheric layer to account for the direct radiative effect in the parameterization.

The expertise of the AC-BSC research team on numerical modeling and its previous experience in the topic makes this task completely affordable.

Task 3.3 Assessment of the radiative forcing of the organic aerosol considering both OA and BrC components

Aerosol radiative forcing is estimated as the difference in the calculated radiative fluxes with all aerosols and with all aerosols except the aerosol type of interest (e.g., OA or BrC). A proper calculation of the radiative forcing of aerosols in models is done by calling twice the radiation scheme in a specific time step of the model. The first call is done considering the effect of aerosols in the radiative balance, and the second call is done without the aerosol type of interest or all aerosols if the total effect is desired. Then, the difference in radiative fluxes provides the radiative forcing of aerosols only. This approach is used in climate models to avoid considering secondary forcings due to the perturbation on cloud formation attributed to the numerical perturbation produced by aerosols far away from their area of influence. This methodology will be implemented in NMMB-MONARCHv1.0. This will allow the model to calculate the instantaneous radiative forcing at specific time-steps or average values over a specific time period (e.g., annual, monthly).

With this detailed methodology implemented, the radiative forcing of all the organic aerosols, both OA and BrC, and their contribution to the total aerosol forcing will be quantified with NMMB-MONARCHv1.0. For that, a global experiment will be prepared based on the configuration defined in Task 1.4b. A 10-year run will be conducted representing present time conditions. Globally and regionally averaged values of the forcing will be quantified, together with different temporal averagings. The internal variability of the forcing will be quantified and compared with current estimates of climate models.

The expertise of the AC-BSC research team in using supercomputer resources assures the success of the numerical experiments planned in this task.

Work package 4: Project management and dissemination of results

The WP4 is devoted to the management, monitoring and dissemination activities.

Task 4.1 Project management

Management of the project will be based mainly in communicating and reporting to the Ministry. Reports have been established periodically one by year. Monitoring of the project is needed for assuring its correct development. Corrective actions will be applied, if required, to reduce deviations from the original plan. Achievement of milestones will provide information on the proper development of the project.

Task 4.2 Dissemination of the project results

The dissemination of the results will be made through: 1) the participation in international conferences, symposia, and other scientifically related activities to present the on going work of the project and discuss with the scientific community about its impact, 2) publication in peer-reviewed journals of high-impact, and 3) the BSC and the Earth Sciences Department web sites. The model developments will be reported in the model webpage. It is planned to provide access to the model code after the end of the project.

C.1.5. Descripción medios materiales, infraestructuras y equipamientos

BSC will provide access to all key research facilities, infrastructure and equipment required for the carrying out of the planned research. BSC hosts MareNostrum4 (Spain's largest supercomputer and one of the six Tier-0 nodes of PRACE) and other HPC specialist machines and research prototypes. MareNostrum 4 has a performance capacity of 13,7 Petaflop/s. The new machine has two distinct parts. The general-purpose element, provided by Lenovo, has 48 racks with more than 3,400 nodes with next generation Intel Xeon processors and a central memory of 390 Terabytes. Its peak power is over 11 Petaflop/s, ten times more than the MareNostrum3. Despite this increase in capacity, it will consume only 30% more power, reaching 1.3 MW/year. The second element of MareNostrum 4 will be formed of clusters of three different technologies that will be added and updated as they become available. These are technologies currently being developed in the US and Japan to accelerate the arrival of the new generation of pre-exascale supercomputers. Like its predecessors, MareNostrum4 will also be connected to the network of European research centers and universities through the RedIris and Geant networks.

C.1.6. Cronograma

Next figure shows the chronogram of the project along the 36 months of the project. Deliverables (D) and Milestones (M) are indicated, and contribution of each member of the group is detailed in the different tasks. Further details are provided bellow, including the list of the work packages, milestones and deliverables specified in the chronogram.

Objectives		1st year												2nd year												3rd year											
	HHRR	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36
WP1																																					
T1.1	PI, PD1, WG2						M1.1						M1.1												M1.1												M1.1
T1.2	RG1, IT1												M1.2																								
T1.3	RG1, PD1, IT1, WG1																	M1.3																			
T1.4	RG3, PD1																																				D1.1
WP2																																					
T2.1	PI, WG3												M2.1																								
T2.2	PI, WG3, WG2																		M2.2																		
T2.3	all																				M2.3																
T2.4	PI, RG1, WG3																																				D2.1
WP3																																					
T3.1	PI, RG2, WG4																								M3.1												M3.1
T3.2	PI, RG2, IT1																													M3.2							
T3.3	PI, RG2, WG4																																				D3.1
WP4																																					
T4.1	PI												D4.1												D4.2												D4.3
T4.2	all												M4.1					M4.1							M4.1					M4.1							M4.1

Principal Investigator (PI): O. Jorba | Research Group: E. DiTomasso (RG1), S. Basart (RG2), F. Benincasa (RG3) | Working group: D. Dabdub (WG1), S. Nizkorodov (WG2), M. Pandolfi (WG3), K. Tsigaridis (WG4) | Hired Personnel: Post-Doc1 (PD1), Informatic Engineer (IT1)

List of Work Packages (description)

WP1. BrC chemistry and role of the reactive uptake of NH3 by SOA: model development and simulations

This work package will be devoted to better understand the chemistry of BrC and the role of NH3 on secondary formation of BrC, first objective of the project. It comprises Task 1.1, 1.2, 1.3 and 1.4 and will be monitored by M1.1, M1.2 and M1.3, and will deliver D1.1. The work package extends the three years of the project, the first year is devoted to literature research and to implement the new chemistry related to BrC. Contributions form Prof. Donald Dabdub and Prof. Sergey Nizkorodov (UCI) are foreseen as experts in BrC chemistry and modeling.

WP2. Experimental campaign

This work package will provide new field measurements to study the BrC chemistry in the northeast Spain, second objective of the project. It comprises Task 2.1, 2.2, 2.3 and 2.4 and will be monitored by M2.1, M2.2 and M2.3, and will deliver D2.1. The work starts in the last part of the first year and extends until the end of the project. Contribution from Dr. Marco Pandolfi (EGAR) is foreseen as expert in optical properties retrievals.

WP3. Radiative forcing of organic aerosols

This work package will assess the role of BrC on the radiative forcing of aerosols, third objective of the project. It comprises Task 3.1, 3.2 and 3.3 and will be monitored by M3.1 and M3.2, and will deliver D3.1. The work starts in the last part of the second year and extends until the end of the project. Contribution from Dr. Kostas Tsigaridis (NASA-GISS) is foreseen as expert in radiative forcing of organic aerosols.

WP4. Project management and dissemination of results

This work package will be devoted to the management, monitoring and dissemination of the project results. It will be feasible thanks to the Project Management Department at the host institution (BSC) and the strong group of “Earth System Services” established at the department level. It comprises Tasks 4.1 and 4.2. Milestones associated to conferences presentation of the ongoing results of the project are foreseen, M4.1. The work package will deliver three annual reports, D4.1, D4.2 and D4.3, with the scientific and economic balance of the project.

List of Deliverables (tracking achievements):
D1.1: Assessment of BrC chemistry and NH3 aging processes on air quality.
D2.1: Overview of results of the summer field campaign.
D3.1: Assessment of the radiative forcing of BrC.
D4.1: First annual report to the Ministry.
D4.2: Second annual report to the Ministry.
D4.3: Third annual report to the Ministry.
List of Milestones (tracking progress):
M1.1: Scientific review of current understanding of BrC chemistry (updated at the end of each year of the project).
M1.2: BrC component implemented in NMMB-MONARCHv1.0.
M1.3: New NH3-SOA chemistry implemented in NMMB-MONARCHv1.0.
M2.1: Compilation of previous field campaigns in Spain devoted to organic aerosols.
M2.2: Design of the summer field campaign.
M2.3: Database of the summer field campaign.
M3.1: Scientific review of the aerosol optical properties (updated at the end of the project).
M3.2: BrC direct effect on radiative scheme of NMMB-MONARCHv1.0 implemented.
M4.1: Presentation in international conferences of the ongoing results of the project.

C.1.7. Contratación de personal
It is foreseen the hiring of a Post-Doc researcher (PD1) and an Engineer Technician (IT1) for the success of the present proposal.

PD1 will work in the design and implementation of the chemistry of BrC in NMMB-MONARCHv1.0 model during two years. PD1 will be directly involved in:
- WP1, Task 1.1: State-of-the-art review of the BrC chemistry and its relevance in ambient conditions and in the troposphere. Furthermore, he will revisit current available schemes of secondary organic aerosol formation suitable for forecasting applications. From the detailed revision conducted, PD1, together with the researchers of the group, will design the schemes to be implemented.
- WP1, Task 1.3: He will work together with the IT1 in the implementation of the new chemistry schemes selected to solve the life cycle of BrC in the NMMB-MONARCHv1.0. Both, primary and secondary origin will be considered along with the aging processes for SOA associated to nitrogen-containing species exposure.
- WP1, Task 1.4: PD1 will be responsible to conduct the numerical experiments to study the role of BrC chemistry on air quality. He will evaluate the surface concentration of organic aerosols, ammonia and ozone, and the aerosol optical depth, absorption and scattering. He will treat data from different measurement networks, e.g., AERONET, satellite data, campaign data.
- WP2, Task 2.2: PD1 will provide technical support during the summer campaign planned for the second year of the project. PD1 will collaborate with EGAR group (in charge of the measurements) in the monitoring of the data obtained.
- WP4, Task 4.2: PD1 will contribute in the dissemination of the results. PD1 will participate in international and national conferences and will be involved in the preparation of scientific publications.

IT1 will work in the programming tasks related with the NMMB-MONARCHv1.0 developments during two years. IT1 will directly work in:
- WP1, Task 1.2: IT1 will code and parallelize the new BrC aerosol scheme within NMMB-MONARCHv1.0. IT1 will evaluate the performance of the new scheme and will improve its parallel efficiency. IT1 will provide support in the tests of the new scheme.
- WP1, Task 1.3: IT1 will program the new scheme of SOA aging due to ammonia exposure. Parallelization and optimization work are foreseen to improve the efficiency of the new code in HPC environments.
- WP2, Task 2.2: Technical support will be provided by IT1 in the monitoring of the air quality and meteorological predictions during the summer campaign planned for the second year of the project.
- WP3, Task 3.2: IT1 will work in the coding of the BrC interaction with the radiation. A deeply review of the aerosol-radiation interaction implemented in NMMB-MONARCHv1.0 will be

conducted by IT1 and the new aerosol component, BrC, will be included in the scheme. IT1 will also implement a new methodology to ease the update of the optical properties of the aerosols in the radiative code.
- WP4, Task 4.2: IT1 will also be involved in the dissemination of the work done in the project. IT1 will present in international and national computing conferences the work done in NMMB-MONARCHv1.0 regarding new parallelization techniques and improvements in parallel efficiency of the system. IT1 will be also involved in publications related with the parallelization and optimization of the NMMB-MONARCHv1.0 model.

C.2. IMPACTO ESPERADO DE LOS RESULTADOS

C.2.1. Impacto científico-técnico, social y económico

Long-term societal impacts of this work will come through an improved understanding of the key precursors and reaction pathways leading to the formation of atmospheric particulate matter. These aerosol particles reduce visibility, have adverse human health effects and impact Earth's climate through their ability to scatter and absorb solar radiation. However, mechanisms by which aerosol particles form and age are complex and still relatively poor understood. In fact, the effect of aerosols remains the largest uncertainty in predicting climate change. SOA accounts for an important but not well quantified fraction of total atmospheric aerosol mass. This work will represent an important step forward in developing next generation atmospheric chemistry models that provide accurate weather and air quality predictions, and allow the key precursors and chemical pathways leading to SOA formation to be identified. This is an essential part of developing effective emissions reduction strategies to mitigate the deleterious effect of aerosol particles on human health and Earth's climate.

The proposed work will result in an updated NMMB-MONARCHv1.0 model that will be immediately available for weather and air quality predictions, as well as for aerosol predictions as part of the International Cooperative for Aerosol Prediction Multi-Model Ensemble (http://icap.atmos.und.edu/) and the CALIOPE Air Quality forecast system for Spain (http://www.bsc.es/caliope/es). These projects directly inform European and international legislators' work to develop air quality policy and regulations. The expected impact of this research, to improve the quality of predictions of aerosol mass and optical properties, and their impact on air quality and climate in one of the premier Spanish chemical weather prediction systems, NMMB-MONARCHv1.0, falls perfectly in line with European research priorities and concurrent initiatives.

C.2.2. Plan de difusión e internacionalización

Dissemination. Results from the proposed work will be disseminated through several channels. First, results will be prepared as a series of scientific articles submitted for publication in GMD, ACP, JGR and/or PNAS. The research group has a history of publishing in these and other high impact journals; Earth Sciences Department of BSC has produced more than 100 scientific publications in the last three years. Additionally, a project web portal will be developed for communication of project information, progress and results to researchers and the general public. The NMMB-MONARCHv1.0 model will be disseminated to the community via the web portal through a versioning control system under development at BSC. Results from the proposed work will be presented at EGU, AGU and other appropriate international conferences. Presentation of the research conducted in the project will be included in the annual BSC Doctoral Symposium, for the PhD-students and post-doctoral researchers involved in the project.

Exploitation. The results from the proposed work will be immediately and straightforwardly exploited for the benefit of European citizens and air-quality and climate researchers globally. First, the updated NMMB-MONARCHv1.0 model will be immediately available for weather and air quality predictions, as well as for aerosol predictions as part of the ICAP-MME and CALIOPE projects. Finally, the results of this work will be made immediately available for exploitation by air quality and climate researchers in several ways:
- The assessment of the potential impact of NH_3 on aerosol mass and optical properties on the global scale will inform funding agencies and laboratory and field researchers as they

allocate resources and propose future research into the complex processes at work in the interactions of NH_3 with gas- and particle-phase species.
- The updated chemistry module made publicly available on the web portal will facilitate the straightforward exploitation of this detailed SOA treatment by researchers in the field as they work to improve SOA predictions in other regional- and global-scale models.
- The NMMB-MONARCHv1.0 chemical weather prediction model will be exploited as the base model for future enhancements, including developing aqueous-phase aerosol and cloud chemistry treatments, for which an accurate, detailed organic and inorganic aerosol module is needed.

Communication of research results and public engagement will be a key focus of the proposed work, and is included in WP4. The research group has a strong history in presentation of research results at scientific conferences, and engagement with the public through publications to the media through the BSC Communication department. Indeed, BSC has dedicated staff and several operational programs in place to communicate activities to other researchers, students, and the general public that will be exploited by the research group as part of WP4. First, important results and milestones will be published in the BSC newsletter for communication to the general public. Existing routes of communication at BSC (website, brochures, presentations, etc.) will also be employed to communicate project information, progress and results. Specific details of the communication through these channels will be developed in conjunction with BSC staff as part of WP4. Members of the research group will further participate in MareNostrum open days, for students and the general public, and professional tours, which exceed 5000 per year. The BSC operates as a PRACE Advanced Training Centre with a mission to provide training and education related to utilization of European supercomputing resources, including for environmental simulation. As part of WP4, members of the research group will participate in the PRACE training program modules related to atmospheric modelling. Finally, results will be presented approximately once a year as part of the BSC Research Seminar Lecture series.

C.2.3. Transferencia de resultados

The results of the project will consist of an improved atmospheric chemistry forecasting system, a better understanding of the radiative forcing of the organic aerosols, and the impact of the direct effect of organic aerosols on meteorological forecasts and climate. In this sense, the transfer of knowledge will be structured through two main plans. First, the improved model NMMB-MONARCHv1.0 will be transfered to the *CALIOPE: Air Quality Forecasting System for Spain* as the main model kernel. The new SOA mechanism and the inclusion of BrC will improve the aerosol predictions of the system. The contribution of BSC to ICAP-MME aerosols global forecasts will be updated with predictions based on the new NMMB-MONARCHv1.0 model. Furthermore, the products obtained with the new aerosol mechanism will be transferred to the World Meteorological Organization (WMO) Sand and Dust Storm Warning Advisory and Assessment System (SDS-WAS) Northern Africa-Middle East-Europe (NA-ME-E) Regional Center and the Barcelona Dust Forecast Center. The centers will distribute internationally the improvements achieved in the model forecast. On the other hand, the scientific outcomes obtained will be disseminated through presentations in international conferences, workshops and publication in international journals. From this dissemination effort contacts may arise with the scientific community to exchange experience and knowledge about the fields of study. Several institutions have expressed interest on the project results: NASA Goddard Institute for Space Studies (NASA-GISS; USA), University of California Irvine (UCI; USA), AEMET Izaña Atmospheric Research Center (Spain), CSIC-Institute of Environmental Assessment and Water Research (Spain), Barcelona Dust Forecast Center (Spain).

C.3. CAPACIDAD FORMATIVA DEL EQUIPO SOLICITANTE

C.3.1. Programa de formación

The training plan envisaged for the Pre-doctoral Contract in the project context will be address to the acquisition of competence in atmospheric modelling and secondary organic aerosol chemistry and radiative forcing. This transition will be ensured by his/her participation in the proposed project and following a training schedule organized by the Earth Sciences Department of BSC that takes advantage of its experience on these issues. The overall

objective of the training program is the transfer of knowledge from the BSC to the Pre-doctoral student in the field of advanced atmospheric and aerosol modelling. The acquisition of expertise in the following scientific topics has been pin-pointed as principal training objective: 1) Meteorology-Air Quality modelling (Emission processes, Transport in the atmosphere, Cloud processes, Dry and wet deposition); and 2) Aerosol radiative forcings (Optical properties of aerosols, Radiative transfer model for SW and LW, Direct effect of aerosols, Meteorological feedbacks). This training will be developed in the framework of the PhD. program in Environmental Engineering at UPC. This Doctorate programme had the MEC Quality Mention until 2010 (MCD2004-00394) and presently has MEC Excellence Mention from 2011 (MEE2011-0335), and it is currently registered in the VERIFICA process of the ANECA evaluation agency (RUCT: 5600080).

The BSC is dedicated to providing high-quality pre-doctoral training that draws upon its experience in developing specific technical and scientific skills, as well as the complementary skills required for efficient research execution and communication. The candidate will have the oportunity to participate on some of the courses organized by the BSC as member of the PRACE consortium. PRACE, the Partnership for Advanced Computing in Europe, appointed BSC as one of the first PRACE Advanced Training Centre (PATC). The mission of the PRACE Advanced Training Centres (PATCs) is to carry out and coordinate training and education activities that enable the European research community to utilise the computational infrastructure available through PRACE. The envisaged courses where the candidate may participate are: PRACE PATC Course Earth Sciences Simulation Environments, and PRACE PATC Course PUMPS Summer School.

The Pre-doctoral Contract will conduct a short-term visit of 4 months with the research group of Prof. Donald Dabdub from the Department of Mechanical and Aerospace Engineering of University of California, Irvine, and the Prof. Sergey Nizkorodov from the Chemistry Department of University of California, Irvine. During this visit, the Pre-doctoral Contract will gain knowledge on the chemistry formation of secondary organic aerosols and its optical transformation when NH3 concentrations are high. Additionally, the Pre-doctoral Contract will contribute to implementing the new chemistry proposed by the group of Prof. Sergey Nizkorodov and Prof. Donald Dabdub in the chemistry model used in the present proposal.

C.3.2. Relación de tesis realizadas o en curso en los últimos 10 años

PhD thesis defended last 10 years:

1) Michele Spada. "Development And Implementation Of A Fully Coupled Global Aerosol Model Within The Chemical Non-Hydrostatic Multiscale Model (NMMB/BSC-CHEM)". 23/11/2015. Spada et al. (2013), Spada et al. (2015).

2) Alba Badia Moragas. "Implementation And Development Of A Gas-Phase Chemical Mechanism Within The Global/Regional Atmospheric Chemical Nonhydrostatic Multiscale Model (NMMB/BSC-CHEM)". 12/12/2014. Badia and Jorba (2014 AE), Badia et al. (2017 GMD).

3) Albert Soret Miravet. "Air Quality Management: Assessing The Impacts Of On-Road Transport Strategies And Industrial Emissions In Urban Areas". 18/12/2014. Soret et al. (2011 APR), Soret et al. (2014 AE).

4) Marc Guevara Vilardell. "Desarrollo De Un Modelo Para La Estimación De Las Emisiones Atmosféricas En España Orientado A La Modelización De La Calidad Del Aire". 17/12/2014. Guevara et al. (2014 AE).

5) Ángel Rincón Rodríguez. "Sistema De Pronóstico De Radiación Solar A Corto Plazo A Partir De Un Modelo Meteorológico Y Técnicas De Post-Proceso Para España". 28/06/2013.

6) Simone Marras. "Variational Multiscale Stabilization Of Finite And Spectral Elements For Dry And Moist Atmospheric Problems". 10/12/2012. Marras et al. (2013ab JCP),

7) Karsten Haustein."Development Of An Atmospheric Modeling System For Regional And Global Mineral Dust Prediction". 31/01/2012. Haustein et al. (2012 ACP).

8) Sara Basart Alpuente."Mineral Dust Model Validation Through Ground Based And Satellite Observation In North Africa And Europe". 30/01/2012. Basart et al. (2009 ACP), Basart et al. (2012 ACP), Basart et al. (2012 Tellus).

9) María Teresa Pay Pérez. "Regional And Urban Evaluation Of An Air Quality Modelling System In The European And Spanish Domains". 22/11/2011. Pay et al. (2010ab AE), Pay et al. (2011 AE), Pay et al. (2012 AE).
10) María Gonçalves Ageitos. "Assesing Variations In Urban Air Quality When Introducing On-Road Traffic Management Strategies By Means Of High-Resolution Modelling. Application To Barcelona And Madrid Urban Areas". 09/03/2009. Gonçalves et al. (2008 AE), Gonçalves et al. (2008 STOTEN), Gonçalves et al. (2009 AE), Gonçalves et al. (2009 STOTEN).

PhD thesis under development:
1) Lluís Vendrell Miquel. "Evaluation And Development Of An Atmospheric Modelling System For A Different Spatial Scales Within The Dust Non-Hydrostatic Multiscale Model: Nmmb/Bsc-Dust". Expected date: October 2017.
2) Vicenzo Obiso. "Aerosol Interaction With Meteorology". Expected date: December 2017. Obiso et al. (2017 JAS).
3) Jaime Pérez Benavides. "Development And Evaluation Of An Air Quality Modelling System Over Barcelona: From Regional To Street Scale". Expected date: July 2019.

C.3.3. Desarrollo científico - profesional de los doctores egresados del equipo de investigación
N/A

C.3.4. Contexto científico-técnico y formativo del equipo y de la institución
The BSC is a public consortium composed of: the Spanish Ministry of Economy, Industry and Competitiveness, the Catalan government and Universitat Politècnica de Catalunya (UPC). The mission of the BSC is to research, develop and manage information technology in order to facilitate scientific progress. The BSC is one of the first eight recipients of the Spanish “Severo Ochoa Centre of Excellence” award given by the Spanish Government, and one of the four host members of the European PRACE Research Infrastructure FP7 project. The BSC hosts the MareNostrum4 supercomputer, used in a Tier-0 PRACE system with 13Pflop/s capacity.

The Earth Sciences Department of BSC (ES-BSC) is focused on carrying out research in Earth system modeling. The high performance capabilities and close collaboration with the Computer Sciences department allow increasing the spatial/temporal resolution of atmospheric modeling systems to improve our knowledge of dynamic patterns of air pollutants in complex terrains and the atmospheric interactions/feedbacks of physico-chemical processes. ES-BSC produces daily operational air quality and mineral dust forecasts for scientific purposes and to support national initiatives for air quality prevention. In addition, ES-BSC is an active air quality model developer, including emission models, aerosol models and chemistry models. Thus, ES-BSC is an appropriate place to conduct the proposed research, that falls within the ES-BSC mission scope, and ES-BSC provides the computational infrastructure required to successfully execute the proposal.

BSC provides professional development plan for each member according to their profile and objectives. In this sense, BSC has been awarded with the Human Resources Excellence in Research due to its progress in aligning their human resources policies with the principles set out in the EU Charter and Code for Research. Additionally, BSC organizes Doctoral Symposiums to allow their PhD students to present and discuss their own research among all the professors and researchers of the center.

C.4. IMPLICACIONES ÉTICAS Y/O DE BIOSEGURIDAD
None.