

## The Fukushima releases: an inverse modelling approach to assess the source term by using gamma dose rate observations.

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A few hour after the earthquake struck the Japan on March 11, the Technical Crisis Centre of the Institut de Radioprotection et de Surete Nucleaire (IRSN), was fully activated, 24/7 for the next 4 weeks to give its expertise to the French government and to the French embassy in Tokyo.

From this experience, lots of difficulties were highlighted. Our consequences assessment capabilities had been limited by uncertainties coming from source term assessment, meteorological data and, on smaller scale, dispersion model.

Since then, the institute has been working on improving its assessment of the atmospheric release and environmental contamination (Mathieu et al., 2012, Korsakissok I. et al, 2013). One of the largest sources of error is the source term estimation including the time evolution of the release rate and its distribution between radioisotopes. Inverse modelling methods have proved to be efficient to assess the source term due to accidental situation (Gudiksen, 1989, Krysta and Bocquet, 2007, Stohl et al 2011, Winiarek et al 2012). Most existing approaches are designed to use air sampling measurements (Winiarek et al, 2012) and some of them use also deposition measurements (Stohl et al, 2012, Winiarek et al, 2013). During the Fukushima accident, such measurements are far less numerous and not as well distributed within Japan than the dose rate measurements. To efficiently document the evolution of the contamination, gamma dose rate measurements were numerous, well distributed within Japan and they offered a high temporal frequency. However, dose rate data results from all the gamma emitters present on the ground and in the atmosphere. A specific methodology based on invert modelling has been developed to operate efficiently dose rate data. Applied to the Fukushima case, the emissions for the 8 main isotopes Xe-133, Cs-134, Cs-136, Cs-137, Ba-137m, I-131, I-132 and Te-132 have been assessed automatically without case specific assumption or guess source term. The Daiichi power plant releases events were well identified and the atmospheric dispersion of the retrieved source term shows a good agreement with environmental observations. The most important outcome of this study is that the method is perfectly suited to crisis management and will improve our diagnosis capabilities in case of a nuclear accident.

Keywords: Inverse modelling, Fukushima, gamma dose rate data, atmospheric dispersion

## Numerical simulation of I-131 concentration emitted from FDNPP using regional model

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Huge amount of radionuclides has been emitted from the Fukushima Daiichi NPP after the 2011 Tohoku earthquake, but the spacio-temporal variation of atmospheric Iodine-131 is still uncertain. Iodine-131 is quite important radionuclides especially for the inhalation, so the reproduction of its spacio-temporal variation is quite useful. For this purpose, we have conducted numerical simulation of Iodine-131 using a regional chemical transport model WRF/Chem which is slightly modified to treat radionuclides' transport, dispersion, and deposition. The emission scenario is taken from the estimation of Chino and Katata. The results shows that the spacial distribution of accumulated concentration of Iodine-131 at the lowest layer of the model is quite different from that of accumulated deposition. The difference seems to be caused by the effect of precipitation, which is quite important removal process from the atmosphere.

Keywords: radionuclides, atmospheric chemistry, material transport, chemical transport model

## JMA's regional ATM calculations for the WMO Task Team on the meteorological analyses for Fukushima Daiichi NPP accident

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The World Meteorological Organization (WMO) convened a small technical task team of experts to produce a set of meteorological analyses that would be used to drive atmospheric transport, dispersion and deposition models (ATMs) for the UN Scientific Committee on the Effects of Atomic Radiation's (UNSCEAR) assessment of the Fukushima Daiichi Nuclear Power Plant accident. The primary aim of the group is to examine how the use of meteorological analyses could improve the ATM calculations.

The Japan Meteorological Agency's regional ATM (JMA RATM) for radionuclides has been developed at the Meteorological Research Institute (MRI), based on the JMA mesoscale tracer transport model for the predictions of oxidant concentration and volcanic ash. The RATM shares its horizontal and vertical grid configurations with the JMA operational nonhydrostatic mesoscale model (NHM) and the JMA operational mesoscale 4D-VAR analysis. With reference to the JMA's global environmental emergency response model, dry deposition, wet scavenging, and gravitational sedimentation for light particles have been revised.

Preliminary and revised calculations of the JMA RATM were conducted according to the task team's agreed standard with a horizontal resolution of 5 km using a unit source emission rate. The simulations were conducted for the period 11 through 31 March 2011. The mesoscale analysis data of JMA were used to drive the ATM, while the JMA's radar/rain gauge-analyzed precipitation data were employed to evaluate the wet scavenging.

Several modifications were made to the JMA RATM. Results of the RATM calculation were verified against the observed Cs-137 deposition pattern and the air concentration time series. The performance of the ATM was significantly improved by the revisions, especially for the Cs-137 deposition.

Keywords: Fukushima Daiichi NPP, Meteorological analyses, WMO Task Team, regional ATM

## Diamond-NICAM-SPRINTARS: simulations on the Fukushima radiation transport

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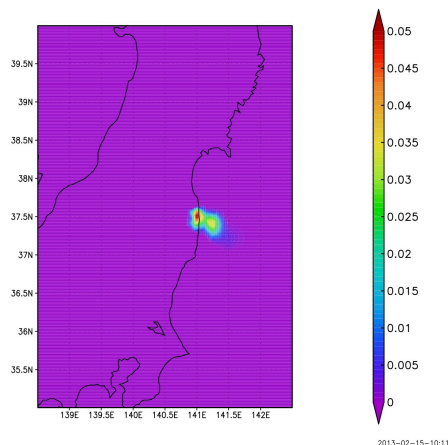
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A Development of Seamless Chemical AssimiLation System and its Application for Atmospheric Environmental Materials (SALSA) project is focused on creating a regional (local) scale assimilation system that can accurately recreate and predict a transport of carbon dioxide and other air pollutants. In this study, a regional model of the next generation global cloud-resolving model NICAM (Non-hydrostatic ICosahedral Atmospheric Model) (Tomita and Satoh, 2004) is developed, which is called a Diamond-NICAM, and ran together with a transport model SPRINTARS (Spectral Radiation Transport Model for Aerosol Species) (Takemura et al, 2000) to simulate aerosols across urban cities (over a Kanto region including metropolitan Tokyo). This enables us to see the effects of warming, pollutants and urbanization to the change in a local climate.

However, for this presentation, we will shift the focus to simulations on the Fukushima radiation transport, recreating a map of radiation spread by analyzing a sulfate transport on the same models(Fig.1). What we are hoping to achieve here is to verify an accuracy of running a finer resolution Diamond-NICAM-SPRINTARS model on transporting an aerosols, as the model is rather new and has not been tested in such occasion. Also, looking at a use of sulfate transport when simulating a radiation transport will be mentioned.

Finally, we will look at and compare a difference between Diamond-NICAM-SPRINTARS and global NICAM-SPRINTARS, and discuss advantages and shortcomings of the models, especially on the accuracy and the time constraints when a finer resolution is applied; also effects of boundary conditions in a regional model is discussed.

Keywords: Diamond-NICAM, Regional model simulations



## The time series analysis of the radionuclide emissions from Fukushima Daiichi nuclear power plant by inverse model

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The accident of the Fukushima Daiichi nuclear power plant that occurred in March 2011 emitted a large amount of radionuclide. The important feature of this accident was that the source position was evidently clear, however, time and vertical emission variations were unknown (in this case, it was also known that the height of emission was not so high in altitude). In such a case, the technique of inverse model was a powerful tool to gain answers to questions; high resolution and more precise analysis by using prior emission information with relatively low computational cost are expected to be obtainable. Tagged simulation results by global aerosol model named MASINGAR (Tanaka et al., 2005) were used; the horizontal resolution was TL319 (about 60 km). Tagged tracers (Cs137) from lowest model layer (surface to 100m) were released every three hours with 1Tg/hr which accumulated daily mean. 50 sites' daily observation data in the world (CTBTO, Ro5, Berkeley, Hoffmann and Taiwan) were collected. The analysis period was 40 days, from 11 March to 19 April. We tested two prior emissions information. The first information was JAEA posterior emission (Chino et al., 2011) and the second was NILU prior emission (not posterior) (Stohl et al., 2012) as our observation data were almost similar to their study. Due to consideration for observation error and space representation error, the observation error was set as 20%. Several sensitivity tests were examined by changing prior emission flux uncertainties. As a result, the prior flux error was set to 100% and JAEA posterior emission flux is used for our analysis. The Cs137 estimated the total emission amount from 11 March to 19 April as 18.5PBq with the uncertainty of 3.6PBq. Moreover, the maximum radio nuclei emission occurred during 15 March, which was larger than JAEA prior information. The results of this study are available for modification of many processes of aerosol transport models. In the future, the combination of regional chemistry transport model and higher time resolution observation data in order to obtain robust emission time series of radionuclide is being planned.

Keywords: Fukushima Daiichi Nuclear Power Plant, Radionuclide, Radionuclide Emissions, Inverse model, Global aerosol model

## Size distributions of airborne radionuclides derived from the Fukushima nuclear accident at several places in Europe

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Segregation and radioactive level analysis of aerosols according to their aerodynamic size were performed in France, Austria, Czech Republic, and Poland after the arrival of contaminated air masses following the nuclear accident at the Fukushima Daiichi nuclear power plant (FDNPP). Sampling was performed from 23rd of March to 23th of April 2011. Whatever the location, the higher activity levels correspond to the finest particle fraction available through Andersen impactors or high volume impactors. As a general pattern, the highest contributions to the <sup>137</sup>Cs and <sup>134</sup>Cs ambient activity level were supported by the finest size range investigated (aerodynamic diameter < 0.49 micrometre). The Activity Median Aerodynamic Diameter (AMAD) ranged from 0.27 to 0.71 micrometre and an average of 0.45 micrometre for cesium isotopes whereas lower values were obtained for <sup>131</sup>I (0.4 micrometre on average). In addition, geometric standard deviation appears to be larger for iodine than for cesium isotopes; probably as a result of gaseous transfer onto ambient particles of all sizes during transport. Contribution from resuspension of formerly deposited <sup>137</sup>Cs was assessed for the coarse particle fractions. AMAD values obtained for natural occurring radionuclides such as <sup>7</sup>Be and <sup>210</sup>Pb ranged from 0.4 to 0.5 micrometre; and 0.3 to 0.4 micrometre, respectively. Larger AMAD values were obtained for <sup>234</sup>Th (3.3 micrometre) and <sup>228</sup>Ac (3.6 micrometre), both radionuclides coming from the resuspension of coarse particles induced by wind erosion of soil surfaces. For the Fukushima-derived fission products, highest weighted activity levels were not found onto the finest particles (< 0.49 micrometre) but in the upper size fraction [0.49 ? 0.95 micrometre] with specific activity levels of about 22 Bq/g for <sup>131</sup>I and around 5 Bq/g for both cesium isotopes. This study also provides a spatial and temporal analysis at the European scale of AMAD values of anthropogenic radionuclides found at significant levels (<sup>134</sup>Cs, <sup>137</sup>Cs, <sup>131</sup>I) as well as for radionuclides of natural origin (<sup>7</sup>Be). <sup>137</sup>Cs and <sup>134</sup>Cs AMAD values at the European scale show spatial homogeneity whereas they gradually decrease from the arrival date to the end of the studied period. This pattern is also observed for <sup>7</sup>Be but with a less pronounced decrease. A specific time trend can be noticed for particulate <sup>131</sup>I (see fig.). Increasing <sup>131</sup>I AMAD values can support the idea of a progressive transfer of gaseous iodine onto ambient aerosols as proposed by Uematsu et al (1988) after the Chernobyl accident or also recently suggested by Mala & Rulik (2013) or Dlugosz-Lisiecka & Bem (2012) after the FDNPP accident.

Figure: Time variation of the AMAD value of Fukushima-labelled aerosols (<sup>134</sup>Cs, <sup>137</sup>Cs, <sup>131</sup>I and <sup>7</sup>Be) in Europe after the arrival of the contaminated air masses (mean values from several sampling sites in Europe).

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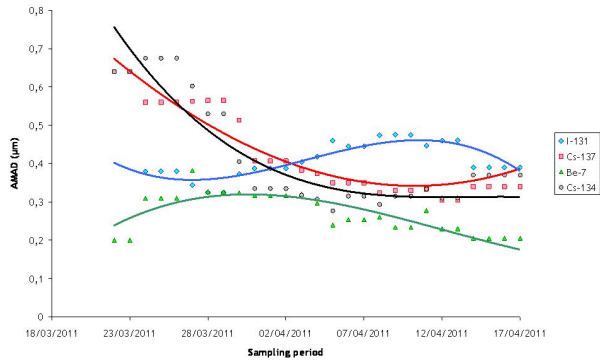
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Keywords: Size distribution, Aerosol, Fukushima, <sup>131</sup>I, <sup>134</sup>Cs, <sup>137</sup>Cs

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## The effect of possible time shift of emission from F1NPP on spatial deposition pattern of $^{137}\text{Cs}$

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Spatial deposition patterns of  $^{137}\text{Cs}$  in east Japan hind-casted by some numerical models so far reported are not always agreed with the monitoring results by the MEXT. There are many unknown processes from the release of radionuclides at the power plant to deposition in the field. In this paper, the source emission estimated by Chino et al. (2011) and by Katata et al. (2012) are shifted at most two hours before and after from the original emission data and results are discussed. The used numerical model is AIST-MM (Kondo et al., 2001), which is a hydrostatic dry-model. The precipitation intensity was introduced with radar-AMeDAS analysis; however, it was not well calibrated due to the destruction of in-situ stations by the earthquake.

The calculated results suggested the earlier release of huge amount of radionuclides in the morning on 15th gave better results.

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Keywords: F1NPP,  $^{137}\text{Cs}$ , Time shift of source emission, deposition pattern



## Evaluation of radioactivity resuspension by mineral dust particles from ground surface using a 1-D vertical model

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Radioactive materials released into the atmosphere by the Fukushima Daiichi Nuclear Power Plant Accident on March 2011 deposited onto various areas such as forest, paddy fields, and so on. The secondary emission of the radioactive materials by strong winds is not elucidated scientifically. This study focuses on the resuspension of the radioactive materials that had adsorbed with mineral dusts from the ground surface, and evaluates the atmospheric radioactivity concentration due to the re-suspended mineral aerosols at the school ground, where was heavily contaminated by the accident, using a 1-D vertical model. The model evaluation was conducted under different soil textures such as sand, loamy soil, silty clay loam and clay. The results were compared with the observation data of the continuously monitored radioactive aerosols at the Tsushima area in Namie town, Fukushima, which evaluated the parameters of the model.

Keywords: the Fukushima Accident, Radioactive aerosol, 1-D model, Resuspension, Mineral dust, Dust emission

## Hourly atmospheric Cs-134 and Cs-137 at SPM monitoring stations in and south of Fukushima after the FD1NPP accident

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No data has been found of continuous monitoring of radioactive materials in the atmosphere in Fukushima area after the Fukushima Daiichi Nuclear Power Plant (FD1NPP) accident on March 11, 2011, although it greatly contributes to accurate evaluation of the internal exposure dose, to reconstruction of emission time series of released radionuclides, and to validation of numerical simulations by atmospheric transport models. Then, we have challenged to retrieve the radioactivity in atmospheric aerosols collected every hour on a filter tape of Suspended Particulate Matter (SPM) monitoring system with beta ray attenuation method used at air pollution monitoring stations in east Japan. A test measurement for hourly atmospheric concentrations of Cs-134 and Cs-137 was successfully performed with a Ge detector for the used filter tapes during March 15-23, 2011, at three stations in Fukushima City 60 km northwest of the FD1NPP and four stations in southwest Ibaraki prefecture 170 km southwest of the FD1NPP. The data in Fukushima City revealed high Cs-137 concentrations of 10-30 Bq m<sup>-3</sup> from the afternoon of March 15 to the early morning of March 16, when a large amount of radioactive materials was simultaneously deposited by precipitation on the land surface according to the measurement of radiation dose rate. Higher Cs-137 concentrations of 10-50 Bq m<sup>-3</sup> were also found from the afternoon of March 20 to the morning of March 21, and which could not be detected by the radiation dose rate due to no precipitation. In contrast, much higher concentrations with the maximum of 300 Bq m<sup>-3</sup> in southwest Ibaraki than in Fukushima City were found on the morning of March 15 and 21 under a strong temperature inversion layer near the surface. The polluted air masses with high radioactive materials were passed away within a few hours as a plume in southwest Ibaraki, while the high Cs-137 concentrations lasted for 10-16 hours in Fukushima City where the polluted air masses after their transport across Abukuma Mountains from the FD1NPP were trapped in the Fukushima basin during the midnight with calm condition. This significant difference in the magnitude of high Cs-137 concentrations and its duration between the two areas was controlled mainly by meso-scale meteorological conditions coupled with topography.

Keywords: Atmospheric Cs-137, Fukushima city, Meteorology, Topography, Time series

## Re-suspension processes of radioactive cesium emitted by the FNDPP accident

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Radionuclides emitted in the Fukushima dai-ichi nuclear power plant (FNDPP) accident have been deposited on the soil, ocean and vegetation. Re-suspension of radioactive cesium from the soil and vegetation to the atmosphere may be one of significant path in the diffusion of radionuclides after the accident.

We have measured the concentration of atmospheric Cs-134/137 radioactivity at several sites in Fukushima region, where deposition amount of Cs-134/137 is relatively high. Atmospheric suspended particle are collected with high/low-volume air samplers mounted at these sites, and gamma-ray emission from them were measured with Ge detector. The measured concentration of atmospheric Cs-134/137 was positively correlated with the wind speed highest during the atmospheric particle sampling period since November 2011, indicating that Cs-134/137 is mainly supplied to the atmosphere by re-suspension with the wind blow. However, contribution of the dispersion of soil particle by the wind is probably limited because variation of atmospheric Cs-134/137 radioactivity concentration mainly occurs in fine particle (diameter < 0.4 micro meter). Assuming that atmospheric Cs-134/137 radioactivity concentration is determined to balance the re-suspension from the surface and the deposition to the surface, we estimated the efficiency coefficients of re-suspension and deposition. We will show factors affecting re-suspension efficiency, and will discuss processes involved in the re-suspension to the atmosphere.

Keywords: Fukushima daiichi nuclear plant accident, environmental radioactivity, re-suspension

## Raidoactive strontium measurement in air dusts using solid-phase extraction

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On March 12, 2011, a large amount of radioactive nuclides (I-131, Cs-134, Cs-137, Sr-90, etc) have been released into the environment by the nuclear accident at the Fukushima Daiichi Nuclear Power Station. There are many radioactivity measurement results of I-131, Cs-134, Cs-137 and the other radio nuclides from air dust collected using high-volume air sampler by germanium semiconductor detector soon after the accident. Activity of these radioactive nuclides in air is important for estimation of internal exposure. However, in these nuclides, some pure beta emitter nuclides have not been measurement sufficiently because of the difficult on chemical isolation for beta ray counting. Strontium-90 is considered one of the harmful radioactive nuclides. Biological half-life of Sr-90 is about 50 years for accumulating in born and the high energy beta rays are emitted from Sr-90 and its daughter nuclide Y-90. Therefore, measurement of Sr-90 is important for calculating exposure. We developed a new strontium isolation technique using solid-phase extraction for determination Sr-90 by liquid scintillation counter (LSC). The technique is simply and quickly for isolating Sr with high-yield.

In this study, we used 3M Empore<sup>TM</sup> Strontium Rad Disk to extract strontium ion from acid solution. However, Pb<sup>2+</sup> ion is also extracted by the disk and Pb-210 interference in radioactive Sr identification. To separate Pb-210, cation exchange is used. 0.02 M EDTA solution can elute Sr ion from the disk. HNO<sub>3</sub> was added to the EDTA solution and passed through cation exchange column with 5% EDTA solution (pH5) to separate Pb. Strontium was eluted by (1+3) HCl. The time of chemical operation for this technique is 3-4 hours without acid treatment and the yield is about 90 %. To determine Sr-90, Cherenkov light of Y-90 has been measured by LSC. With sequential measurement, the growth curve of Y-90 was described to determinate activity of Sr-90.

We measured Sr-90 in air dust samples of Hitachi, Kawasaki and Osaka. The result of Hitachi is shown in Fig.1. In Hitachi, the Sr-90 activity concentration in air is decreased with time and the ratio of Sr-90/Cs-137 is about 10<sup>-3</sup>. It is possible that after April, Sr-90 has been the same behavior of Cs-137. We will discuss about time variation of radionuclides activity of Sr-90 and proportion of Sr-90 to Cs-137.

Sampling date	<sup>90</sup> Sr Bq/m <sup>3</sup>	error %	<sup>90</sup> Sr Bq/ <sup>137</sup> Cs Bq	error %
2011/4/9	1.5E-03	2.5%	1.6E-03	10.3%
2011/4/18	3.7E-04	2.4%	1.1E-03	10.3%
2011/5/21	6.3E-05	4.7%	3.7E-03	11.0%

Fig.1. The results of Hitachi air dust samples

## Atmospheric-Terrestrial Interactions in Radioactive Pollution by the Fukushima Accident

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Our Grants-in-aids Scientific Research Group have been established to elucidate the physico-chemical state of the radioactive aerosols that were released into the atmosphere by the Fukushima accident (primary emission), improve transport model by which better emission inventory is made, and understand better the atmospheric-terrestrial interactions in the radioactive pollution. While aiming at providing basic data on the estimation of the inhalation exposure, as well as the establishment of simulation model for radionuclides transfer among atmosphere-terrestrial-hydrosphere, the final goal in the project is to contribute and ensure the safety as well as security of the people by providing relevant information on the internal exposure due to atmospheric radioactivity. This project promotes the state-of-the art methodologies from various research fields to be integrated with the comprehensive research methods/frameworks, and has specific sets of topics as listed below.

1) Monitor continuously radioactive aerosols and gases in the atmosphere around the Fukushima Daiichi Nuclear Power Plant, particularly at the greatly contaminated regions with higher radiation dose rate, to examine temporal change in concentration.

2) Analyze samples that have been collected soon after the accident in order to understand the physico-chemical properties of the radioactive aerosols, detailed radionuclide composition, particle chemical composition and particle size distribution, as well as correlation with specific stable isotopes.

3) Observe the atmospheric radioactivity concentration under various conditions, as well as the suspension of soil particles and the aerosol particles along with meteorological elements such as soil water content, by making an observation field in the polluted area mentioned above. Furthermore, perform a necessary controlled experiment and better understand the re-suspension process (secondary emission) of the radioactive material from the soil. Apply the up-to-date mass spectrometry on this occasion, with measuring directly radionuclides and elements in atmospheric aerosols to clarify their relations with meteorological parameters.

4) Quantify the emission flux of radionuclides, which is supplied by eco-processes such as the suspension of Japanese cedar pollen and wax-like substance on the plant surface (secondary emission).

5) Verify the present assumption of the simulation model concerning wet deposition process of radionuclide by comparing observation results of the radioactivity in air and in the precipitation.

6) Validate the simulation results by comparison with those of each observation activity as well as improve the quantification in transport, diffusion and deposition in the atmospheric models, finally to develop resuspension model to quantify the atmosphere-terrestrial interactions.

In this presentation, the briefing of the project is given with expecting active discussion among attendees; what kind of atmospheric science should be indispensable.

Keywords: the Fukushima Accident, Radioactive aerosol, Primary emission, Secondary emission, Resuspension